NUCLEAR SAFETY

A REVIEW OF RECENT DEVELOPMENTS

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foreword

This quarterly journal is a Technical Progress Review prepared by the Oak Ridge National Laboratory at the request of the Office of Technical Information, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the field of nuclear safety. *Nuclear Safety* is not a comprehensive abstract of all literature published in this field during a given quarter; rather it is a mechanism for presenting concise reviews of selected subjects as prevailing interest and available information warrant.

Coverage of the Review is limited to topics relevant to the analysis and control of hazards associated with nuclear reactors, operations involving fissionable materials, and the products of nuclear fission. Primary emphasis is on safety in reactor design, construction, and operation; however, safety considerations in reactor fuel fabrication, spent-fuel processing, nuclear waste disposal, and related operations are also treated. Safety in the use of radioisotopes in industry, medicine, and research is excluded, as are most topics considered to be in the province of health physics. Even with these exclusions, nuclear safety overlaps such diverse fields as nuclear physics, solid-state physics, mechanics, chemistry, meteorology, geology, seismology, metallurgy, law, and nearly all branches of engineering. The authors will therefore review material from these fields which, in their opinion, has a direct bearing on nuclear safety.

Three distinctly different types of articles are in this issue of *Nuclear Safety*: reviews of current literature, special review articles on specific topics, and comparative studies of various aspects of U. S. power reactors. The editors feel that each of these articles makes a necessary and distinctive contribution to this journal. The special review articles permit discussion of pertinent subjects which cannot be adequately considered by reference to only the current literature. The comparative studies direct attention to acceptable practice in the U. S. power-reactor industry. The current review articles, however, constitute the major portion of this issue.

All incoming literature (including reports, books, American and foreign technical journals, and transactions) is examined for subjects within our area of interest. This material is collected, grouped, and reviewed by experts. Interpretations, in any article, represent the opinions of the editors, who are employees of the Oak Ridge National Laboratory. Readers are urged to consult references to original work for more complete information.

In addition to the invited contributors, many members of the Oak Ridge National Laboratory staff wrote review material, reviewed manuscripts, or otherwise contributed to this publication. Their contributions are gratefully acknowledged.

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Section I

GENERAL SAFETY CRITERIA

Safety Practices in Western Europe

It has become a recognized axiom that the expected or potential gains from nuclear energy utilization must be evaluated in comparison with the estimated risks of physical and financial damage. The potential costs and requirements for coordination of technical knowledge, developments, and skills are of such magnitudes that, in much of the world, only governments or associations of governments are able to take leading roles in nuclear energy activities. Whether development is carried out by private or public capital, the minimization of risk to the public, consistent with maximum gain for the public, is recognized as a basic requirement that must be evaluated and regulated by governmental bodies. In the past several years a number of documents have appeared which reflect this concern in Western Europe. The documents have originated from both individual countries and the international organizations that deal with atomic energy.

Protection of Personnel from Ionizing Radiations

Most countries are well aware that the entry of private industry into the field of nuclear energy production requires the enactment of legislation defining the rules to which nuclear installations must conform and establishing who shall be liable with respect to emitted radiations. Great Britain passed a major milestone in this regard with the introduction of the Nuclear Installations (Licensing and Insurance) Bill[1] by the Government in the House of Lords toward the end of 1958. The resulting law[2] was put in force in April 1960. Briefly, the United Kingdom law provides that:

1. All nuclear plants must be licensed.
2. The nuclear site license is given for a site and an operator who will be a corporate body, and not to an individual.
3. Conditions may be attached to the license, and the license can be revoked at any time.
4. An authorization for the discharge of waste is necessary.
5. The length of the period during which a licensee is legally responsible can be altered by the Minister of Power.
6. The licensee is absolutely responsible.
7. Claims for injury or damage must be submitted within 30 years.
8. Claims made after 10 years will not be paid until Parliament decides where the money is to come from.
9. Insurance or reserves up to 5 million pounds (sometimes more) must be provided.
10. There can be collective cover where one licensee has three or more sites.

Several countries of Western Europe have been guided toward legalistic measures for the control of nuclear and radiological installations by the European Atomic Energy Community (Euratom) Commission. The first step in this guidance was the publication of the basic standards[3] to which member states should adhere. These standards provided the separate national authorities with a common basis for safety legislation and thus constituted the first step toward the establishment of uniform safety standards in the Euratom countries (Belgium, France, Germany, Italy, Luxembourg, and the Netherlands). Drafts of numerous laws and regulations were subsequently submitted to the Euratom Commission for over-all coordination. The status of these is given in the following statements quoted from a recent Euratom report:[4]

Belgium

On 6 March 1958, the text of Draft Law No. 385 on the protection of the population against ionizing radiations was submitted to the [Euratom] Commission for its opinion in accordance with the terms of Article 33. The Commission conveyed to the Belgian Government its favourable opinion.

On 29 December 1959, the Commission received a draft Royal Decree regulating the possession and use of radioactive substances for medical purposes, on which it likewise gave a favourable opinion.

A set of preliminary draft general regulations on the protection of the population against ionizing radiations has also been communicated to the Commission for information purposes.

Germany

On 29 October 1958, the Commission received the Federal Government's draft Atom Law (Atomgesetz). The Commission gave a favourable opinion.

On 22 January 1960, it received the text of the draft Decree on Radiation Protection (Strahlenschutzverordnung). The Commission's comments have been forwarded to the Federal [German] Government.

France

The Basic Standards are currently being studied with a view to their application by the various ministries concerned. No text has been submitted to the Commission by the French Government to date.

Italy

On 12 January 1960, two preliminary draft sets of safety standards for the prevention of hazards arising from ionizing radiations (one drawn up by the Ministry of Health and the other by the Ministry of the Interior) were transmitted to the Commission for information purposes. The two drafts are broadly in line with the provisions of the Euratom Basic Standards. Other texts, adopted before the entry into force of the Euratom Treaty, have also been submitted by the Italian Government.

Luxembourg

On 31 March 1958, the Commission received the text of a preliminary draft law on the protection of the population against the dangers arising from ionizing radiations. It has conveyed to the Luxembourg Government its favourable opinion.

Netherlands

On 5 February 1960, the Commission received the text of the draft nuclear law submitted to the Second Chamber of the States-General. The text is now being studied.

On 12 February 1960, a set of draft regulations on radiation protection was forwarded to the Commission for information, and is now being studied.

Other European countries are taking similar legalistic action under the aegis of the Organization for European Economic Cooperation (OEEC); and, in August 1959, all 17 OEEC nations (Euratom countries plus Austria, Denmark, Greece, Iceland, Ireland, Norway, Portugal, Sweden, Switzerland, Turkey, and the United Kingdom) agreed on common measures to safeguard populations from radiation. The OEEC Council adopted a resolution requiring member countries to ensure adequate health protection for all persons who might be exposed, whether occupationally or otherwise.[5] All countries were requested to report to the European Nuclear Energy Agency on measures taken by November 1959.

Reactor Safety Evaluation

Many countries, including those mentioned above, have introduced measures requiring that the installation of a reactor be permitted only after proper certification of safety by a competent authority. Since such certification must necessarily be based on the judgment of experts and the pooling of expert knowledge, which is not always readily available to individual countries, the International Atomic Energy Agency (IAEA) has taken steps to provide a reactor safety evaluation service.[6] Certain general procedures have been adopted for carrying out assignments of this type.

At the request of a member state for a reactor safety review, an evaluation group, consisting of members of the IAEA scientific staff, first examines and evaluates a detailed report dealing with all aspects of the proposed installation which might have a bearing on safety. The report is prepared by those in charge of the design and construction of the reactor. On the basis of this evaluation, the system is revised as necessary by the designer, and the report is prepared for presentation to an advisory panel on reactor safety. On the basis of the opinion of this panel of experts of international repute, the national authorities decide on the safety certification of the reactor. This procedure is followed before the construction of the reactor and again before the actual operation of the reactor.

The procedures will vary, of course, to some extent according to the needs of individual cases, but a good working model was established in the course of the first reactor safety evaluation

carried out by the IAEA. This was done at the request of the Swiss Government for the Swiss reactor project "Diorit." The IAEA not only obtained the services of the experts for the final evaluation but it also advised the reactor staff on the preparation of the report on the safety analysis and on its presentation to the experts. The advisory panel concluded at the end of its examination that the reactor could be operated without undue risk to the health and safety of the public, and, accordingly, the panel submitted a report to the Federal Council of Switzerland.

The Swiss authorities have expressed their appreciation of the work done by the panel and of the assistance given by the IAEA at various stages of the evaluation. The Swiss authorities have also requested the IAEA to arrange for similar evaluations of power-reactor projects in Switzerland, and arrangements for these are already under way. In addition, a few other governments have begun inquiries regarding the possibility of IAEA assistance in this field, and it is possible that the evaluation of reactor safety will some day become one of the major activities of the IAEA. A valuable contribution to the efforts to solve the technical problems concerned with reactor safety will be made by panels of experts which the IAEA plans to convene for discussing questions of siting and containment of reactors.

In countries that have qualified personnel, the safety clearance of reactors is carried out on an individual basis. At Harwell, for example,[7] the Research Group Reactor Safety Clearance Committee has the duty of assessing the safety of reactors (including subcritical assemblies) in Research Group establishments. This Committee operates primarily through working parties (one for each reactor) and, based on considerations of the findings of the working party, gives or does not give safety clearance. The working party is composed of members who have no executive responsibility for the reactor and have between them the required breadth of experience, including up-to-date familiarity with reactors of the type concerned. Findings are based on examination in great detail of a safety document prepared by those responsible for the design and/or operation of the facility. A safety clearance is valid for a limited period and is subject to periodic review.

Site Selection Criteria

The criteria used for selecting a site for a nuclear power reactor or for a nuclear research establishment differ in the various foreign countries, depending on individual requirements and conditions. In Spain, for example, the choice of sites for two large-scale nuclear plants now being considered for construction was based on economic considerations rather than on safety considerations.[8] In the case of the Madrid site, the first step in the selection was to locate sites with suitable amounts of cooling water (40 m^3/sec) within a radius of 100 km from Madrid. The only satisfactory site from this standpoint was found to be 85 km from Madrid and 20 km from Toledo. A plot of population density as a function of distance from the site was then compared with similar plots for the nuclear plants at Dresden, Shippingport, and Bradwell. The results indicated a population density comparable to that in the vicinity of the Dresden plant and considerably lower than that around Shippingport or Bradwell. With this low population density and favorable meteorological, hydrological, and seismological conditions, it was concluded that the selected site would be acceptable. No mention was made in the reference report of the need for a gastight building for reactor containment. A similar approach was taken in selecting the Seville site.

As in the case of the Madrid and Seville sites, the selection of Mol as the site for the Belgium Nuclear Research Center was also based on factors other than safety.[9] Although it was realized that the Mol site was in a region of relatively high population density (at the time of the selection in 1955, 112,000 persons lived within a radius of 15 km, and 575,000 persons lived within a radius of 30 km), the site was chosen because of its availability; soil conditions; water supply; accessibility by road, railroad, and canal; and the availability of nonspecialized personnel nearby. Construction of the BR-2 high-flux research reactor and the BR-3 power reactor at Mol was not foreseen; however, it was later decided that suitable precautions could be incorporated into the design of these two facilities to ensure the safety of the people living in the vicinity of Mol. These precautions consisted primarily of enclosing the reactors in pressuretight buildings and providing radioactivity monitoring stations to cover all directions around each of the facilities concerned.

Because of the experience at Windscale, the British are very cognizant of the importance of applying safety criteria in selecting sites for nuclear power stations; however, conventional engineering requirements (e.g., foundations, access, and water supply) are normally the governing factors.[10,11] In each case, careful consideration is given to the factors leading to the release of radiation during normal operation and under accident conditions, and the plant is designed so that the neighboring public outside the site boundary is not normally exposed to levels of radiation above the limit of 0.5 rem/year, as recommended by the International Commission on Radiological Protection (ICRP). According to Farmer and Fletcher,[10] this ICRP recommended limit does not impose any undue restrictions on the choice of sites for the large gas-cooled power reactors, because radiation in the vicinity of such a reactor can be held below the limit by proper design without larger exclusion radii than would otherwise be required for a satisfactory power-plant layout.

Although factors connected with the normal nuclear operations do not usually limit the choice of site, the consequences of various accidents that could be caused by the failure of equipment or by maloperation of the plant can be very restrictive. Since in the Windscale incident attention was drawn to gaps in the knowledge of the biological effects of different levels of exposure, this matter has been given careful consideration by the British Medical Research Council.[12] As a result, emergency dose limits have been recommended which can be used to assess the merits of a particular reactor site relative to the consequences of a reactor accident. The Council recommends that the maximum daily intake of I^{131} attributable to an accident and in the period following it should not exceed a total thyroid irradiation of 25 rads, and the intake of Sr^{89} and Sr^{90} should not exceed a total of 15 rads and an annual rate of 1.5 rads/year, respectively, at sites of highest concentrations in bone. The implication of these limits from the standpoint of preventing excessive doses occurring through inhalation, food consumption, and whole-body irradiation is illustrated below, based on I^{131}. The effects of all other iodine isotopes, as well as strontium and the radioactive gases krypton and xenon, have been taken into account, where appropriate, in arriving at the levels quoted.[10]

Inhalation. A hazard arises from the inhalation of radioactive iodine by persons exposed for a few hours to the passing cloud. With the recommended standards, it is estimated that the time-integrated airborne concentration of I^{131} should be limited to 0.01 curie-sec/m^3.

Consumption of Exposed Foodstuffs. The recommended limit on daily intake is 0.3 μc for young persons (ages 3 to 20). This corresponds to a ground contamination of 5×10^{-5} curie of I^{131} per square meter and assumes that each person ingests an amount of radioactivity per day equivalent to that from 60 cm^2. A separate milk restriction limit that applies to young children has been recommended as 0.065 μc/liter. By the use of the data obtained after the Windscale accident, this can be related to a herbage contamination level of approximately 0.5 μc of I^{131} per square meter. As the iodine decays, the milk restriction limit will be reduced to that imposed by Sr^{90}, 0.002 μc/liter.

Whole-Body Radiation. A limiting distance is determined at which *ground* contamination would give rise to an integrated whole-body dose of 25 rem. The contamination level to meet this case is 2×10^{-2} curie of I^{131} per square meter. If 25 rem of whole-body radiation is received through exposure in the *cloud*, the equivalent dose to the thyroid through the inhalation of iodine would be 5000 rem. In this case, clearly, the thyroid dose is controlling, and the whole-body radiation limit is irrelevant.

The British Medical Research Council emergency standards normally applied to the selection of a site in the United Kingdom are as follows:[10] "(a) Few people should live within 500 yards of the site. (b) In any 10° sector around the site, not more than 500 people should live within 1½ miles. (c) No large centre of population of 10,000 or more should lie within 5 miles." These standards would have the following effects in the event of a ground release of 250 curies of total iodine containing 110 curies of I^{131}. People living within 440 yards of the site would be evacuated temporarily, and there would be a temporary ban on the consumption of crops and exposed food. There would also be a temporary ban on the consumption of milk for people within a radius of 4 miles from the site. For a release ten times larger, the radius for temporary evacuation would be increased to 1500 yards, the radius for tem-

porary ban on crops and exposed foods would increase to 1 mile, and the temporary ban on milk consumption would be applied within 14 miles of the site.

The influence of safety criteria on the selection of sites for nuclear installations in countries other than those cited previously varies widely, depending on local conditions. In Switzerland, for example, almost all sites with satisfactory water-cooling capabilities are within 10 km of settlements with 10,000 inhabitants or more.[13] Thus the possibility of applying exclusion-area criteria to the choice of sites in Switzerland is remote. For this reason, attention has turned to underground installations close to large towns. Such installations have the economic advantage that they can be used for combined heat and power production; they also have several advantages with respect to safety.

The cost of an underground nuclear plant in Switzerland was estimated to be about 2.5 per cent more than the cost of the same plant constructed aboveground, excluding the cost of the land in each case. After maintenance costs for the steel shell of a power station aboveground were considered, as well as the higher capital outlay resulting from the purchase of the necessary land, it was concluded that a power station aboveground would probably not be cheaper than an underground station. Similar considerations have lead to the construction of underground nuclear plants in Sweden and Norway.

Safety criteria used in selecting sites for nuclear plants and research installations in Italy, as in the other Euratom countries, are comparable to those used in the United States.[14–16]

Safety and Control of Transported Nuclear Materials

The public safety aspects of transporting fissionable materials and radioactive materials constitute a special problem in many foreign countries because of high population densities, the need for transporting material from country to country, and the lack of uniform safety standards. At the present time an international convention exists concerning the carriage of goods by rail, and an annex to this sets out the conditions for transporting radioactive materials on international rail routes. Packages are exempt from these regulations if they contain less than specified amounts of radioactive material and if the dose rate on the surface is less than 10 mr per 24 hr. The International Air Transport Association (IATA) Regulations relating to the carriage of restricted articles include a section on radioactive materials; and, finally, the Merchant Shipping (Dangerous Goods) Rules, 1952, detail requirements on the carriage by ship of toxic materials and explosives (to which class radioactive materials belong). At the present time, however, observance of all these regulations is far from uniform.

The IAEA is attacking this problem, and draft regulations are being prepared on the transport of radioactive substances by land, sea, and air.[17] Two panels have been set up to draft recommendations in this regard, with the task to be completed in 1960. One panel is dealing with the transportation of radioisotopes and radioactive ores and residues of low specific activity, and the other panel is dealing with the transportation of large radioactive sources and fissile material. In the future, it is anticipated that the IAEA recommendations may be universally accepted, and much greater uniformity will be achieved on an international scale.

In the United Kingdom the transportation of radioactive materials on both a national and an international scale is controlled by regulations that have been in existence for a number of years; however, these regulations have recently been revised and are now awaiting ministerial approval.[18] Methods for protecting transport employees in the United Kingdom from the hazards associated with transporting radioactive materials are based on the ICRP recommendations for maximum permissible cumulative doses to the whole body for nonoccupationally exposed persons of Group B (special groups).[19] The annual dose is thus limited to 0.5 rem, which is one-tenth the permitted average exposure for "occupationally" exposed persons. This high level of protection to the transport employees is achieved by enforcement of specific criteria[20] for packaging, monitoring, and handling radioactive materials.

Conclusions

It is evident from the foregoing that many countries, particularly those of Western Europe, are cognizant of the public safety problems associated with the construction of nuclear installations or the transportation of radioactive isotopes and other nuclear materials and are taking

legal measures to ensure that persons in nonoccupational, as well as occupational, categories are fully protected. The Euratom Commission, the OEEC, and the IAEA are all working toward the establishment of uniform safety regulations and the acceptance of these on an international basis. (J. A. Lane)

First Report of the Federal Radiation Council

The first report[20] of the Federal Radiation Council (FRC) is of more than ordinary interest because it gives the initial indication of the role this new governmental body may assume in the field of radiation protection. In the past, the various governmental agencies, as well as other users of radiation, have tended to follow the recommendations of such unofficial groups as the National Committee on Radiation Protection (NCRP), the American Standards Association (ASA), or the International Commission on Radiological Protection (ICRP). The FRC was formed in 1959 (Public Law 86-373) to "... advise the President with respect to radiation matters, directly or indirectly affecting health, including guidance for all Federal agencies in the formulation of radiation standards and in the establishment and execution of programs of cooperation with the states ..." The full report may be considered to be of a summary and advisory nature, but a memorandum embodying the principal conclusions of the report was endorsed by the President and printed in the *Federal Register* of May 18, 1960, and it thus became the official policy of the federal government.

The actual conclusions reached in the document do not present any marked differences from recommendations of such groups as the NCRP. Nevertheless, there are some changes of emphasis which are worth noting. Section I is introductory and gives a brief résumé of the formation of the various official and nonofficial groups that preceded the FRC in this field. The introduction also limits the scope of the report to what might be termed normal peacetime operations involving nonmedical uses of man-made sources. Thus, emergency situations arising from accidents are not considered.

Sections II and III of the FRC report give a brief summary of general information concerning radiation effects in man and on the present levels of background radiation from natural sources, as well as from fallout and other man-made sources. Although there is no bibliography detailing the sources of this information, these sections do provide a very brief summary of the more important conclusions that seem plausible on the basis of current data or theories.

Section IV outlines the considerations to be used in setting radiation protection standards. Since a threshold dose below which radiation effects do not exist has not been demonstrated and seems rather unlikely by most theories of radiation damage, any exposure must be assumed to entail some risk of injury. Thus, any exposure is justified only on the basis that certain benefits are expected from the application, and radiation protection involves the balancing of the expected risks and benefits. The risk can be estimated only by extrapolation to low dose levels of the incidence of effects observed at higher levels or by comparisons with the exposure due to natural background or to industrial experience at these exposure levels. The latter sources of information give essentially negative results since no valid epidemiological study has been made which could establish an incidence rate of the order of the natural incidence of most comparable effects, such as leukemia and bone cancer. The report does not indicate any method of estimating the benefits or how these might be compared meaningfully with the risks.

Section V is devoted to the basic guides. The FRC has rejected the terminology of "maximum permissible doses" and "maximum permissible concentrations" because it is believed that these terms have acquired a connotation of a sharp distinction of a level below which one is "safe" and beyond which there is "hazard." Therefore, the terms "Radiation Protection Guide" (RPG) and "Radioactive Concentration Guide" (RCG) are adopted to avoid any connotation of change in the response curve at this level. The definition of the RPG is rather vague and cannot well be taken out of context. Admittedly, the inferred break in response is not inherent in the terms, or at least is not so interpreted in the case of other such operational guides (e.g., maximum speed), and it will be interesting to observe whether the new terms, by application, will acquire much the same connotation as the former. RPG values (in rem, as formerly) are given for occupational exposure, as well as for exposure of the gonads or the total body in the

case of population exposure. In all cases, values are identical with those recommended by the ICRP[21] and the NCRP.[22] This report does not give the RPG values for single-organ exposure in the case of population exposure, but it indicates that these may be the subject of a later report.

Section VI gives general consideration to the establishment of the RCG values, although none are explicitly recommended in this report.

Section VII is a summary of the report. In particular, Paragraphs 7.7 and 7.13 reemphasize that, since an RPG involves a balancing of benefits and risks, "there is no single 'permissible' or 'acceptable' level of exposure without regard to the reasons for permitting the exposure. The Guides may be exceeded only after the Federal agency having jurisidiction over the matter has carefully considered the reason for doing so in the light of the recommendations in this report." This latter sentence occasioned considerable comment during the hearings of the Joint Committee on Atomic Energy in May and June of this year.[23] The interested reader may wish to refer to the discussion and testimony relative to this point.

The recommendation that the agency involved may decide, after careful consideration, to exceed the RPG recommended in FRC Report 1 is of particular interest because one of the criticisms, which perhaps led to the formation of the FRC, was to the effect that the agency which was interested in promoting a particular application was also charged with adopting regulations and standards for the protection of employees and/or the public. In the testimony at the Congressional hearings,[23] representatives of the FRC indicated that the actions of several federal agencies would be reviewed by the FRC. This could alter greatly the force of these recommendations. Although this may resolve the question at the federal level, the testimony at the Congressional hearings indicated that there are sources under the control of states, communities, or even private companies and individuals which need to be considered.

Although FRC Report 1 cannot be said to have given a final answer to these perplexing questions, it does indicate that some progress has been made. For the first time, the federal policy on matters of radiation protection is being determined. The policy remains under scrutiny at the highest level of the government because the FRC is essentially a group at the Cabinet level. The very existence of such a body may be more significant than any particular pronouncement of this report since the real effect often will be apparent only as it is applied.

(W. S. Snyder)

Fuel-Element Handling

Fuel handling has caused incidents that resulted in serious contamination of equipment and buildings with attendant loss of operating time and exposure of personnel during cleanup. Fuel meltdown or contamination from ruptured elements not only can cause trouble but mistakes in storage or shipment can lead to criticality. Such accidents must be avoided. This review is concerned specifically with a number of recent reports dealing with loading fuel elements into reactors, unloading them, and storing them after irradiation. Information on the shipment of spent fuel elements was presented in the September 1960 issue of this Review.[24]

Loading and Unloading

Loading fuel into a reactor is not especially hazardous if the reactor is not operating. Care must be taken, however, not to damage the fuel elements because this could result in the subsequent loss of fission products. During the unloading of a reactor, there may be additional problems associated either with the removal of afterheat or the contamination from a ruptured fuel element. Removal equipment must be designed to control the potential contamination from a ruptured fuel element. Provisions must be made for keeping the fuel-element temperature well below the melting point of any constituent during the unloading process.

Gas-Cooled Reactor at Berkeley, England. The power reactor at Berkeley, England, will have a unique method[25] of unloading and reloading during operation, for which highly specialized machines have been developed. Since the purpose of this reactor is to produce electricity as continuously as possible, the ability to load and unload while operating is especially important. The designers elected not to use multipurpose machines but, rather, to have specialized equipment for individual jobs wherever possible. The aim was simplicity of operation, which should reduce operational mistakes and mechanical failures. The design includes a

chute-handling machine that will be used to install and remove the chute through which fuel elements will be removed and charged. Separate machines will be used for charging and discharging fuel elements. All three pieces of equipment will have electrical and mechanical interlocks to ensure the proper sequence of operation. All interlocks have been designed on a "fail-safe" basis. Because the entire operation must be conducted while the reactor is at full power, provisions are included to contain the CO_2 used under pressure to cool the reactor. Each machine can be considered a pressure vessel. The apparatus for discharging fuel has a circulating cleansing and cooling system that normally employs air but which will use CO_2 while connected to the reactor.

Organic-Moderated Reactor at Piqua, Ohio. A system for the installation and removal of reactor fuel has been designed by Atomics International for an organic-moderated power reactor that will be located at Piqua, Ohio.[26] An extensive mockup was built in order to conduct both "dry" and "wet" runs for removal of fuel elements and control rods. During wet runs, the fuel elements were handled under liquid, unirradiated Santowax R (the organic moderator material) at temperatures ranging from 300 to 650°F. This type of mockup testing is imperative for systems that do not provide direct visual control of the operation since complete reliance must be placed on indexing, positioning, load- and position-indicating devices, and interlocks. Chances for failures of any of the equipment must be minimized.

"Analysis showed that two hours after shutdown a fuel element which was removed from 350°F organic fluid would heat up from decay heat generation and melt the aluminum cladding in 19.6 minutes. In the average handling cycle the fuel element was out of the liquid organic about five minutes before immersion in the water pool."[26] In approximately 1 min after the water has been hooked up, it is possible to water-cool the fuel element in the cask. Careful time and motion analyses were made to optimize the time required to complete a fuel-handling cycle. The time was approximately 27 min. As a result of these mockup tests, a list of 24 detailed pertinent recommendations, ranging from small mechanical changes to operational modifications, has been derived.

At the Organic-Moderated Reactor Experiment, a combination fuel removal and shipping cask has been used successfully.[27] "The basic cask is two feet in diameter by eight feet in length and weighs 16,000 lbs." During the loading of the cask, an auxiliary shield extension is moved into the empty section of the reactor tank. Approximately 18 in. of unshielded space exists, however, between the extension shield and the organic liquid. As a hot fuel element is pulled from the liquid into the auxiliary shield, a radiation field of 10 r/hr exists at a radial distance of 30 in. from the upper reactor flange. During this same short period, the operators would be working in a 200 mr/hr radiation field.

Sodium-Cooled Reactor of the USS Seawolf. During January and February 1959, the first unloading of fuel from the S2G reactor[28] (the USS *Seawolf's* sodium-cooled reactor) was successfully completed. The job was done outdoors in freezing weather. "Operations were severely handicapped during one 3-day period because of winds with velocities of 40 to 50 mph with gusts to 75 mph. These winds were accompanied by temperatures from 0 to 15°F. In general, winds below 20 mph caused no difficulties. Winds above 40 mph caused complete cessation of crane operations. These high winds were from the west and had relatively little effect on the motion of the *Seawolf.* During one [other] 3-day period, southeast winds were encountered and swells and vessel motion did develop."[28]

Many minor difficulties were encountered, but they were solved quickly and efficiently so that the operation continued on schedule. In fact, the actual transfer time per fuel element was closer to 30 min than to the estimated 1 hr. After the transfer had been completed, sodium was cleaned from the fuel elements, and the fuel elements were shipped in casks to Idaho for reprocessing.

Storage

The storage of irradiated fuel elements is an important problem at both operating and processing sites. Facilities for cooling, monitoring, decontaminating, and controlling criticality are usually required.

Eurochemic has proposed a processing plant with a capacity of 350 kg of uranium a day.[29] The metal will be received in casks weighing up to 70 tons. Before the carriers are unloaded,

water will be pumped through them, and the effluent will be checked for radiation to detect ruptured fuel elements. Provisions will be made for decontamination of the casks before they are returned to the reactor. Inactive ends of fuel elements will be removed by shearing. Active material will not normally be exposed in this operation. If irradiated fuel is accidentally exposed during removal of the fuel ends, it will be canned immediately to prevent contamination of the water. The storage ponds will be arranged to prevent the spread of contamination and will be equipped with a demineralization system. The floors are to be stainless-steel-covered, the walls will be liquid tiles or their equivalent, and the underwater tools and equipment will be of stainless steel. In each case the reason for the special material is to make decontamination as easy as possible.

The "ideal" storage canal has been described as follows:[30]

1. A method for selectively withdrawing settled solids from the bottom of the canal is needed. The canal should slope towards a central low point; and a method for flushing the solids to this area would be desirable.
2. A cleanup system consisting of a pump, filter for particulate matter removal, and a demineralizer unit is necessary.
3. A method for dissolving these solids, once they have been collected, for recycle to processing plants should be available; or a method for removal to the burial grounds would suffice.
4. The canal wall surface should be smooth enough to permit cleaning. The use of stainless steel or ceramic tile would be ideal; however, the cost may be prohibitive. The use of various sealing compounds should be investigated.
5. Ample freeboard between the surface of the water and the top of the canal wall should be provided.

Spent fuel will be stored in a clean water pool at the Experimental Gas-Cooled Reactor (EGCR), as outlined for the preliminary Partially Enriched Gas-Cooled Power Reactor (PEGCPR) design.[31] Known ruptured elements will be received in a small compartment through which pool water will be purged to prevent the release of fission products. After the ruptured elements have cooled so that they are safe to handle, they will be canned. As fuel is received, each piece will be inspected for defects, and records will be made. This information will be quite valuable to reactor operations personnel and to fuel manufacturers. (W. R. Casto)

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Section II

ACCIDENT ANALYSIS

Chemical Reactions Involving Liquids

Chemical reactions of liquid fuels or liquid coolants, as well as reactions which would transform solid reactor components into liquids, are important to consider from the nuclear safety standpoint. Several examples of such reactions which conceivably could cause or contribute to nuclear accidents, in either a reactor or a radiochemical plant, are discussed here, but only a few of the many types of reactions involving liquids that may be significant in nuclear safety are considered. In most of the cases cited, a complete understanding of the possible hazard requires a knowledge of the phase behavior and, in particular, detailed phase diagrams of the materials which may undergo reaction, as well as reliable information on reaction rates. Proof of the stability of a set of materials under planned operating conditions provides no guarantee that unpredicted variations from those conditions will not initiate a hazardous reaction. The determination of the detailed phase behavior of the materials is often experimentally difficult, time-consuming, and expensive, but the high stakes involved in nuclear safety and successful operation of both reactors and radiochemical plants would seem to make such comprehensive studies mandatory.

Formation of Liquid by Solid-Phase Reactions

Eutectic melting between two metals in contact in a nuclear reactor may occur with disastrous consequences if the solidus temperature is exceeded. Neymark[1] studied the reaction of fuels considered for the Sodium Reactor Experiment (SRE) with their thin-walled stainless-steel jackets in order to determine whether the rate of eutectic formation was rapid enough (<1 min) to cause ultimate reactor shutdown if an extensive temperature excursion occurred. Machined fuel specimens ($\frac{3}{8}$ in. square and $\frac{1}{8}$ in. thick) were balanced in a quartz tube on larger squares cut from type 304 stainless-steel tubing with a wall thickness averaging 0.0105 in. The quartz tube was evacuated, inserted in a preheated furnace, and removed after a few minutes for examination.

In the experiments,[1] unalloyed uranium and a uranium alloy containing 3.5 wt.% molybdenum were found to react with the steel specimens relatively slowly below the melting point of the fuel (~2000°F). At 1800°F, from 4 to 10 min was required for penetration of the steel; at 2200°F, however, penetration or melt-through took place in less than 1 min. In experiments with a thorium alloy containing 8 wt.% uranium (melting point, ~2900°F), penetration or alloying occurred at 2300°F after 2 to 2.5 min, and there was some alloying after 4.38 min at 2000°F. It has been mentioned by Bentle[2] that the thorium-uranium phase diagram shows the presence of a small amount (1 to 7 per cent) of liquid phase for this alloy composition between 1965 and 2210°F. Apparently this small quantity of liquid does not reduce the time required for melt-through to less than 1 min.

There was concern with respect to the SRE fuel-element reaction rates because of certain postulated high-temperature, short-lived reactor transients. Although data concerning the effects of such transients were obtained, it subsequently developed that there was an unanticipated mechanism for obtaining high fuel-element temperatures for long periods of time. Thus, although the data[1] seemed to indicate that fuel-element melting would not occur, this did not prove to be the case.[3] Neymark[1] properly stated, however, that the intimacy of contact

and surface cleanliness should profoundly affect the reaction rate. In the experiments reported, thin oxide films, which should increase melt-through time, were present on the fuel specimens. The questions here are: How much reliability can be placed on thin oxide coatings for slowing down such reactions in all cases, and what is the effect of an intense radiation field on these coatings?

Reaction of Liquid Metals with Water

Since the reaction of liquid sodium-potassium alloy with water is highly exothermic, is usually very rapid, and produces hydrogen gas and caustic alkali, it is necessary to know whether there is danger of a serious accident if a reactor condition exists in which this reaction can take place as a result of a leak. In connection with the planned irradiation of several uranium rods in NaK-filled aluminum capsules in a water-cooled reactor at Hanford, Weber[4] carried out tests in which capsules were deliberately made to fail in a water environment in order to determine the consequences in event of a capsule failure.

Several NaK-filled aluminum capsules were fabricated for the tests, and each contained a Wood's metal plug driven into a $\frac{1}{16}$-in. hole. The Wood's metal was tested to melt at 76°C. Three capsules that were heated in water in an autoclave at 170°C for 2 hr showed no distortion and only a trace of external corrosion around the holes, but there was severe internal corrosion. One of the capsules continued to leak a highly basic liquid for about 15 min after removal from the autoclave. Four capsules were held for 2 hr in a reactor-size flow tube with recirculating water at 93°C, 60 psi, and a flow rate of 25 gal/min, and another four capsules were tested under the same conditions for 18 hr. The flow-tube tests also produced no distortion. There was external corrosion but very little internal corrosion, indicating that the flowing water had immediately flushed NaK and the reaction products out of the capsules.

The results of the tests indicated that, in the event of a capsule failure, there would be no distortion which might injure the reactor process tube. The NaK-water reaction is so potentially hazardous that each reactor experiment in which this reaction is a possibility should, of course, be previously studied under simulated conditions.

Stability of Organic Coolants

Mixtures of polynuclear organic compounds have been considered for use as reactor coolants because of their almost complete lack of corrosiveness and because, in contrast to aqueous coolants, low-pressure operation is possible with them. The hazards associated with radiolytic decomposition and combustion of such compounds were discussed in a previous issue of this Review.[5] An additional safety consideration is the stability of these mixtures with respect to phase segregation during reactor operations.

Desirable characteristics for an organic coolant mixture are described in a paper by Troutner.[6] It is stated that, in addition to good pyrolytic and radiolytic stability, the mixture should have a high boiling point, should be liquid at 25°C, and the vapor in equilibrium with the liquid should be of such composition that it is liquid when condensed at 25°C. The last characteristic stems from the recognition that solids condensing on the walls would not melt back into the liquid and that the resulting composition change would drastically alter the properties of the coolant.

Troutner[6] determined melting-point diagrams and obtained vapor composition data for binary and ternary mixtures of biphenyl, *o*-terphenyl, *m*-terphenyl, naphthalene, and phenanthrene. He defined an optimum mixture as that with the lowest melting point at which the vapor composition is liquid when condensed at the liquidus temperature. Melting point in this case refers to the liquidus rather than the solidus temperature. It was found that no combination of the five compounds formed optimum mixtures that melted below 35°C. As an example, the eutectic in the *o*-terphenyl–biphenyl system contains 57.5 mole % *o*-terphenyl and melts at 29.5°C. The optimum mixture contains 78 per cent *o*-terphenyl, and the liquidus temperature is 46°C. Several of the ternary systems have lower eutectic temperatures and optimum mixtures. All parts of a reactor coolant system must be maintained at temperatures above the optimum temperature of the particular organic mixture chosen as the coolant.

Reactions of Molten-Salt Fuels

The Aircraft Reactor Experiment (ARE) used a circulating molten mixture of UF_4, NaF, and ZrF_4 as fuel and primary coolant, and a fuel

containing LiF, BeF_2, UF_4, and ThF_4 is to be used for the Molten-Salt Reactor Experiment (MSRE). One of the safety considerations in choosing a fuel composition is that the solution must be stable with respect to precipitation of a uranium-rich phase in event of small composition variations, contamination of the solution, or a temperature drop as a result of reactor malfunction.

The phase diagram of the $NaF-ZrF_4-UF_4$ system[7] is very complex, containing six $NaF-ZrF_4$ and five $NaF-UF_4$ binary compounds. The primary phases over most of the diagram are solid solutions between UF_4 and ZrF_4, $7NaF \cdot 6UF_4$ and $7NaF \cdot 6ZrF_4$, and $3NaF \cdot UF_4$ and $3NaF \cdot ZrF_4$. In the $LiF-BeF_2-UF_4$ system[8] the primary phases over most of the diagram are uranium-containing compounds. The availability of detailed diagrams for these and other pertinent systems permits a confident choice of a composition which will not move into a primary phase field of a uranium-rich phase upon small composition variations or will not precipitate such a phase upon inadvertent cooling to below the liquidus temperature.

Contamination of a molten-fluoride fuel solution with oxygen or an oxygen compound is a potential hazard because insoluble UO_2 will be precipitated.[9] In the proposed MSRE, there are two ways in which such contamination can occur: (1) by incomplete oxygen removal during fuel preparation or during pretreatment of the graphite moderator with which the fuel will be in contact during reactor operation and (2) by accidental introduction of oxygen during maintenance operations such as replacing pumps or heat exchangers. Since oxygen or its compounds react with ZrF_4 in preference to UF_4, inclusion of ZrF_4 in the fuel solution is being considered.

Another type of reaction that may be hazardous can occur if certain ZrF_4-bearing fuels are cooled to below the solidus temperature in a reactor, heat exchanger, or dump tank.[10] The fuel, 57 mole % NaF–42 mole % ZrF_4 –1 mole % UF_4, for example, which was once among those considered for a molten-salt power reactor, might behave in a hazardous manner. This fuel is nearly a eutectic mixture (40.5 mole % ZrF_4) in the $NaF-ZrF_4$ system.[7] Upon cooling, the eutectic mixture freezes at 500°C, and a few degrees lower the compound $3NaF \cdot 2ZrF_4$ is often produced by a solid-phase reaction between the eutectic components, $2NaF \cdot ZrF_4$ and $7NaF \cdot 6ZrF_4$. When this reaction takes place, there is an increase in the bulk volume of the solid. It is not known whether rupture of a large containing vessel would occur upon freezing of a fuel with a composition similar to this particular eutectic composition, but the possibility would have to be considered.

Phase Reactions of High-Temperature Aqueous Fuels

The Homogeneous Reactor Experiment (HRE-2) uses a circulating fuel consisting of a dilute solution of UO_2SO_4, $CuSO_4$, and D_2SO_4 in D_2O. Because of a slow reaction of D_2SO_4 with stainless steel, $NiSO_4$ appears in solution and usually must be considered as a fuel component. The core temperature of the HRE-2 averages about 240 to 280°C. There is a pressurizer joined to the fuel system which contains D_2O that is held generally at a temperature above 300°C. Reactions involving phase changes of the fuel mixture have particular significance for safe reactor operation since it is very important that the fuel solution in all parts of the reactor system be stable toward precipitation of uranium-containing phases which might be swept into a region of high neutron importance.

Marshall and Gill[11] have studied solid-liquid equilibria in the five-component system at 300°C. Solubility data for $NiSO_4 \cdot H_2O$, $UO_3 \cdot nH_2O$, and the hydrolytic solid compounds $2UO_3 \cdot SO_3 \cdot 5H_2O$, $CuO \cdot 3UO_3$, and $3CuO \cdot SO_3 \cdot 2H_2O$ were obtained at 0.06*M* SO_3 concentration. There is a negative temperature coefficient of solubility for the hydrolytic solids. Concentrations of HRE-2 fuel solutions are far enough away from the solubility boundaries that precipitation of solids could not occur at 300°C or below in the reactor core.

At the junction of the pressurizer and fuel system, however, there is an increased temperature, as well as dilution of the fuel with D_2O, which decreases the acidity. Both factors increase the likelihood of hydrolytic solid precipitation. Indeed, at this location in both the HRT mockup and after HRT run No. 7 with depleted fuel, crystalline solids were found which were identified[12,13] at a later date as essentially $CuO \cdot 3UO_3$.

Another type of phase reaction of aqueous fuels is separation into two liquid phases when the temperature is increased[14] to the range 300 to 370°C. One of the phases, referred to as the heavy liquid, is highly concentrated in

uranium and begins to settle out of the main body of solution as soon as the liquid-liquid immiscibility temperature is reached. A theory advanced to explain the power excursions in the HRE-2 is that the heavy liquid, produced in the system by one of several possible mechanisms, accumulates in a region of low neutron importance at the bottom of the core vessel and is then occasionally swept into the region of high neutron importance.

One mechanism postulated to explain how the heavy liquid could be produced originally is that local overheating above the two-liquid-phase temperature of the fuel (~332°C) is initiated by hot spots on the core wall produced by absorbed uranium compounds or by particles of uranium-containing solids swept through the core. Another postulated mechanism is that, at limited locations in the core, the fuel is concentrated at a temperature below that of the system pressurizer, and, after several-fold concentration, the two-liquid-phase temperature falls below the fuel temperature. The heavy liquid then precipitates.

Two ORNL reports[15,16] on liquid-liquid boundary temperatures show that the two-liquid-phase temperature of one HRE-2 fuel drops from 332°C to a flat *minimum* of 305°C upon concentrating between 6 and 16 times and that increasing the amount of excess acid raises the minimum temperatures. A theory discussed in the reports is that if the pressurizer temperature is held below the minimum temperature, concentration of the fuel cannot result in formation of the heavy-liquid phase and consequent reactor operational instability. Subsequent reactor experiments have indicated semiquantitative agreement with this notion.[17]

(R. E. Moore)

Reaction of Uranium in Bromine Trifluoride Solutions

In an effort to reduce the cost of nuclear power and to more efficiently utilize natural resources, considerable effort is being directed toward processes for recovering uranium from spent fuel. One such process, known as the fluoride-volatility process,[18] involves the conversion of uranium fuel to uranium hexafluoride by direct fluorination with liquid bromine trifluoride. This reaction is exothermic, releasing 1500 cal per gram of uranium. The exact mechanism of this reaction is unknown, and, as a result of the fluoride-volatility pilot-plant accident[19] at Brookhaven National Laboratory (BNL) on May 15, 1957, studies were undertaken to determine the effect of certain variables on this reaction. The incident was discussed in more detail in the September 1960 issue of *Nuclear Safety*.[20]

At the time of BNL pilot-plant accident, five unirradiated slugs (1.1 in. in diameter and 4.0 in. long) were being dissolved at 121°C in a BrF_3 solution containing 4.4 mole % UF_6, 4.8 mole % BrF_5, and <0.1 mole % Br_2. After approximately 3½ hr of apparently normal dissolution, five explosions occurred in rapid sequence and resulted in extensive damage to the equipment. The most probable cause of the accident was believed at that time to be the ignition of a partially dissolved slug when it was lifted into the vapor section of the dissolver by the slug height probe.

In more recent studies[21] at BNL, uranium specimens of various geometries have been exposed to solutions containing controlled amounts of BrF_3, BrF_5, and UF_6 in a reaction chamber where the temperature and pressure were controlled. Ignition occurred in both the liquid and vapor phases. Although ignition occurred at a lower temperature in the vapor phase, 118°C as compared with 325°C (metal temperature) in the liquid, detonation was not observed, presumably because limited quantities of the reactants were present. Several runs were made with oxide-coated uranium milling chips which had an average specific area of 85 cm^2/g. Immediate ignition was observed in all runs with a solution temperature of 21 to 30°C. Detonations occurred in several runs, and the detonation pressures were in excess of 12,000 psi.

The increased hazard that exists when there are confined areas in the uranium being dissolved was demonstrated. Small cubes, ¼ in. on each side, were stacked in both random and close-packed arrays. Although ignition occurred in both cases while submerged in solution at temperatures of approximately 150°C, severe detonations were observed when the specimens were randomly stacked. Similar behavior can be expected when the confined areas resulting from pitting of the uranium slugs develop during the dissolution process. Such areas may be difficult to control since the rate of chemical attack is dependent upon numerous variables, such as

distribution of impurities in the uranium, composition of solution, and temperature.

The surface temperatures of several small uranium cylinders were recorded during dissolution and were found to oscillate, with peak values as high as 1100°C. This indicates a surface film reaction of considerable potential violence in a sustained reaction. At normal dissolution temperatures the conversion of uranium to UF_6 probably proceeds in a stepwise manner, as evidenced by the catalytic effect of UF_6 and Br_2. The rate-controlling step is presumably eliminated at sufficiently high temperatures during detonation.

The experiments at BNL clearly point out the hazardous nature of the dissolution of uranium in UF_6-BrF_3 solutions, especially since submerged detonations were obtained under conditions likely to be encountered during full-sized slug dissolution. The role of each of the many variables of such a process has not been clearly defined, and the importance of additional work is evident. (H. E. McCoy)

Interactions Involving Graphite in Nuclear Reactors

Graphite has long been in use as a thermal-reactor moderator. Its attractiveness is a result not only of its moderating properties, but also of its availability, low cost, good machinability, and moderate chemical reactivity. However, the push for nuclear power systems having greater efficiencies has increased operating temperatures to the point that the reaction rate of graphite with other essential components in the reactor core becomes important. In view of the difficulties encountered in servicing or replacing the moderator during the life of a reactor, a thorough study must be made of possible compatibility problems prior to construction of a system utilizing graphite. In a review article by Carter,[22] a survey of the characteristics of the possible interactions of graphite with other core materials was made. The following discussion is based on Carter's article and on other pertinent literature.

Graphite and Coolants

Water has been used as a cooling medium in a large number of graphite-moderated reactors, with the coolant and the moderator being separated by metal tubing. In the event of a failure of the separating material, saturation of the graphite with water would occur and would pose two primary problems: (1) the macroscopic neutron absorption cross section of the graphite would increase about 10-fold, and (2), if the graphite temperature were above 600°C, the water-gas reaction

$$C + H_2O \rightarrow CO + H_2$$

would result in rapid oxidation of the graphite.[23] The water could be removed from the graphite by conventional drying techniques, but the large bulk of the material and its porosity would make the process somewhat tedious. The use of high-density graphite might greatly reduce the problem.

Liquid sodium and sodium-potassium alloy have also been employed as coolants in graphite-moderated reactors. Again, the coolant and the moderator were separated. Failure of the separating material would not, however, result in a violent chemical reaction. Sodium, like potassium, is believed to form an interlamellar compound with graphite in which layers of metal atoms are inserted between layers of carbon atoms.[24–28] Although the formation of this compound causes dilation of the carbon axis of the graphite, evacuation at a pressure and temperature which allow vapor transport results in complete removal of the metal and recovery of the graphite dimensions. No serious cracking of the graphite occurs as a result of the anisotropic expansion due to sodium absorption. However, the presence of more than 200 ppm potassium in the sodium leads to decrepitation. The presence of both sodium and potassium increases the neutron absorption cross section of graphite.

From the standpoint of chemical reactivity, helium is an obvious choice for a gaseous coolant in a graphite-moderated system. Helium is an inert gas, and hence only the effect of the trace impurities present must be considered. On the other hand, the thermodynamic and nuclear properties of carbon dioxide make it an attractive coolant, and it is compatible with graphite at temperatures below about 700°C in the absence of radiation.[29] In the presence of ionizing radiation, incompatibility is observed[30] at temperatures in excess of 500°C. Above these temperatures, significant quantities of carbon monoxide are generated by the reaction

$$CO_2 + C \rightarrow 2CO$$

This reaction can result in the movement of large amounts of graphite from the higher temperature regions of the reactor core to cooler regions of the system.

Hydrogen is an attractive coolant from the standpoints of availability, thermal-energy transport, and momentum transfer. Although one of the primary problems associated with the use of hydrogen as a coolant is its explosive reaction with oxidizing media, it also poses some problemes with regard to the graphite moderator. Several gaseous hydrocarbons are formed above 600°C, and ionizing radiation has been shown to cause such species to be formed[31] at 500°C. For irradiations of the order of 2×10^{18} thermal neutrons/cm^2, the hydrocarbon partial pressure rises toward 0.2 per cent of the hydrogen pressure. Hence, if the hydrocarbons formed are continually removed from the gas stream, either by reaction with other reactor components or by purification, serious loss of the moderator may occur.

The problems associated with the use of organic liquid coolants in contact with graphite are quite interesting. Because of gamma and neutron heating and the poor conditions for heat transfer, the liquid absorbed into pores of the graphite will be at temperatures above those of the coolant stream. If this process proceeds at a reasonably slow rate, the graphite is converted to a high-density, partially graphitized solid. If the reaction rate is extremely high, physical distortion of the graphite will occur. Again, these problems are somewhat alleviated by the use of high-density graphite.

The use of air as a coolant involves the compatibility of graphite with nitrogen, oxygen, water, carbon dioxide, argon, and other gases that occur as reaction products. Several of these components, namely, carbon dioxide, oxygen, and water, are exothermically oxidizing to graphite. Hence the use of air as a coolant at elevated temperatures is not feasible. Serious damage may also result in systems utilizing other gaseous coolants if a large amount of air inleakage occurs during operation.

Graphite and Structural or Cladding Materials

Several reactors have utilized aluminum as a fuel-cladding material. Aluminum and carbon are nonreactive at temperatures slightly less than the melting point of aluminum;[32] however, reactions may occur between aluminum and impurities which may be desorbed from the graphite. These impurities are principally carbon monoxide, carbon dioxide, and water, which are rapidly reduced by clean aluminum until a protective layer of aluminum oxide limits the reaction rate.

Stainless steel has been used widely as a reactor structural material. The transport of elemental carbon into stainless steel can occur at elevated temperatures; however, carburization is most likely to occur as a result of contact with some intermediate carbon-bearing medium. This medium may be a gaseous coolant containing small amounts of carbon monoxide or methane. Liquid sodium in contact with graphite has also been demonstrated to be a potent carburizing medium[33] above 500°C. Moderate carburization is not serious because the strength of the stainless-steel component is increased without appreciable loss in ductility. However, gross carburization under certain thermal and mechanical conditions greatly reduces the rupture ductility of the material.

Zirconium and Zircaloy have not as yet posed any serious problems, since their physical properties have discouraged their use at temperatures above 600°C. At temperatures above 800°C, the diffusion rate of elemental carbon into these materials becomes appreciable, and a carbide case is formed. This layer is protective unless oxygen is present, in which case the carbide layer flakes. Hence an unpredictable amount of corrosion can occur as long as the environment is oxidizing. The reaction of zirconium with hydrogen is also detrimental. If the hydrogen concentration becomes high enough, a hydride phase precipitates, leading to embrittlement of the metal.

A series of alloys, called Magnox alloys, which contain magnesium, beryllium, aluminum, and other trace elements, has been employed in power reactors in the United Kingdom. These alloys are reported to be unreactive to carbon and dry carbon dioxide up to their melting points.

Coatings for graphite offer a potential solution to many of the graphite compatibility problems. Numerous coatings that have been investigated show various degrees of protection. If the purpose of the coating is to prevent erosion of the graphite, the permeability of the coating is not a problem, and several coatings

are available for this application. If the coating is needed to prevent some medium from penetrating the graphite, then the coating must be impermeable. Small openings seem invariably to occur in most of the coatings, with siliconized silicon carbide being a notable exception. In an oxidizing environment, carbon oxides that are formed at the base of a flaw in the coating rise to the surface, where *in situ* formation of SiC and SiO_2 results. These compounds tend to seal the flaws in the coating.

Graphite and Fuel or Moderating Materials

Uranium metal and graphite react[34] to form carbides at temperatures above 1500°C. Below 1200°C the reaction rate is negligible in the absence of some medium for carbon transport. The carbides formed have higher melting points than uranium, and their presence does not result in any adverse change in the physical and mechanical properties of uranium. Uranium oxide is likewise stable in contact with graphite until temperatures in excess of 1700°C are reached.[35]

Under certain conditions it may be desirable to use both BeO and graphite as moderator materials in the same reactor. Reactions between graphite and BeO do not occur at an appreciable rate[36] at temperatures less than 2300°C. In other systems, beryllium metal may be used in the core with graphite serving as the reflector. Beryllium-carbon reactions do not occur rapidly until temperatures approaching the softening point of beryllium are reached. However, any carbon monoxide evolved from the graphite reacts with beryllium to form a surface layer of carbon and powdery BeO. The finely dispersed state of the BeO combined with its toxicity makes it a serious potential hazard.

Because of the good moderating properties of hydrogen, the hydrides of several metals which have low nuclear cross sections are being investigated for use as moderators. These include the hydrides of magnesium, yttrium, zirconium, columbium, and cesium. If carbon-bearing species such as carbon monoxide or CH_4 are present and the metal hydride is at 900°C or above, the hydride may be decomposed by the following reaction:

Carbon (CO, CH_4, etc.) + metal hydride

→ metal carbide + hydrogen

This reaction will result in serious alteration of moderator properties if the temperature and environmental conditions remain favorable for its occurrence.

Graphite and Control-System Poisons

Boron compounds are widely used for the control of nuclear systems. It is conceivable that extreme temperature excursions may result in the formation of boron carbides. These compounds would present no significant hazards since they would remain where they were formed until mechanically removed. The use of boron trifluoride is also anticipated. If this compound comes in contact with graphite, it will react with metallic impurities in the graphite to form volatile fluoride compounds. A local deposit of boron or boron carbide, depending upon the temperature, would result, and its presence would cause a severe rise in the macroscopic neutron absorption cross section of the moderator graphite.

Cadmium is also an attractive control material, but its primary disadvantage is its volatility. Above 500°C, cadmium is volatile and is easily transported to other parts of the system. This element also grossly affects the neutron absorption characteristics of the moderator.

(H. E. McCoy)

Pyrophoricity of Uranium and Zirconium

Under suitable conditions uranium and zirconium exhibit chemical reactivity that is shown in pyrophoric behavior and oxidation reactions of explosive violence. There have been numerous incidents in which uranium chips ignited during machining operations. A large number of massive pieces, ½ in. thick, were once ignited when a truck was involved in a motor vehicle accident.[37] The fire lasted nearly 24 hr and could not be extinguished. The Windscale incident[38] in England, although initiated by the release of stored energy in graphite, is an example of the hazards of uranium burning in a nuclear reactor. Although no one was seriously injured, the resulting contamination of surrounding countryside, the unfavorable publicity, and the damage to the reactor testify to the seriousness of the incident.

Numerous incidents involving the ignition and detonation of zirconium are also recorded. At Bettis Atomic Power Laboratory, 159,000 lb of zirconium scrap stored in open segregated bins ignited for no apparent reason.[39] The fire was so intensely hot that windows over 100 ft away were cracked. In another incident, at the Y-12 Plant salvage yard at Oak Ridge, two men were killed and a third lost his arm when a drum of finely divided zirconium metal exploded.[40]

The pyrophoric potential of metal powder is well known, but, as the above incidents illustrate, massive pieces of zirconium and uranium can also exhibit pyrophoric behavior. An exothermic reaction is a necessary, but not sufficient, condition for pyrophoricity. Physical and chemical couplings between the reacting material and the environment determine the rate of energy generation and the rate of energy dissipation. Hence particle size and heat-transfer properties are important factors, with higher specific areas and lower heat-transfer rates favoring pyrophoric behavior. The release of products from the primary oxidation reaction which are capable of exothermic reaction with the environment is also a contributing factor. The combination of the factors mentioned and several others not discussed can lead to reactions that vary from a slow, almost isothermal reaction to an explosive release of reaction energy, with resultant detonation and shock waves.

As an illustration of the temperatures attainable by the ignition of metals, Conway and Grosse[41] have calculated the adiabatic combustion temperature for the Al-O_2 reaction to be 3500°C at 1 atm pressure. Doyle et al.[42] have calculated the Zr-O_2 reaction temperature to be about 4650°C, and there are indications that this temperature was attained in an experimental torch.

Reactions of this nature are important considerations in reactor construction, because zirconium and uranium are frequently used in a system where potential oxidants are present in the coolant or moderator. The use of sensitizers, such as aluminum, in contact with zirconium and uranium is also commonplace. The proposal of more exotic reactor concepts and the desire for higher temperatures make it imperative that more be learned about the pyrophoric properties of metals and of the variables which control pyrophoricity.

An excellent review of the present knowledge of the pyrophoricity of uranium has been written by Zima.[43] In light of the information disclosed in his review, several specific suggestions were made with regard to the Hanford reactors. These involve steps for early detection of cladding defects and minimization of time required for the removal of defective elements. It is also recommended that a quench gas, such as helium or carbon dioxide, or an inert foam blanket be available for regular or standby use during discharge operations.

The Argonne National Laboratory is currently involved in a research program concerning the ignition behavior and kinetics of oxidation of uranium, zirconium, plutonium, thorium, and binary alloys of each. Two modes of attack are being used in their studies. In a direct approach, several different types of ignition experiments have been developed in order to study important factors such as metal purity, particular alloying additions, specific area, and composition of the oxidizing gas. Since ignition is basically oxidation, a fundamental study is being made of the isothermal oxidation characteristics of these materials. A status report[44] of this program was published last year. The studies of thorium and plutonium were still in the planning stages at that time, but several interesting results concerning uranium and zirconium were presented. These results are summarized in Table II-1 essentially as presented by Schnizlein et al.[44]

This discussion serves to point out the potential hazards that exist in some reactor systems because of the pyrophoricity of uranium and zirconium components. The ignition or detonation of such materials could not only cause serious damage to the reactor but could also endanger operating personnel through fire and explosion hazards and/or release of fission gas from the reactor. The multitude of factors which control reactions of this type have been pointed out, and quantitative information has been presented relative to some of these factors. However, many factors remain unexplained, and much additional work will be required to resolve the present problems, as well as additional ones posed by new reactor concepts. (H. E. McCoy)

Table II-1 COMPARISONS OF OXIDATION AND IGNITION BEHAVIOR OF URANIUM AND ZIRCONIUM[a]

	Uranium	Zirconium
	Incidents	
Form of metal involved	Powder, machine chips, and frequently bulk metal	Usually powder and machine chips, rarely bulk metal
Initial temperature	Frequently heated by fire, fission products, or machining	Usually ambient temperature
Violence	Rarely explosive	Frequently explosive
	Ignition Behavior Investigations	
Methods applied	Burning curves[b] and shielded experiments[c]	Shielded experiments[c]
Forms of metal used	Bulk and foils	Foils
Atmospheres studied	Air[d] and oxygen[e]	Air and oxygen
Ignition temperatures		
Observed for 1-mil foil (pure metal)	335°C[f]	640°C
Additive effects		
Bulk samples	355–780°C[f,g]	
5-mil foils	380°C[f,h]	530–800°C[i]
Additive causing greatest decrease	Al, Nb, and Si (bulk samples only)[f,g]	Ti and Al[i]
Additive causing greatest increase	Cu (bulk samples only)[f,g]	Ni and Cu[i]
Correlation to isothermal data	None evident	Reciprocal to weight gain in first minute at 700°C[i]
Specific area dependence, where T_{ign} = ignition temp., °K, A = area, cm², m = mass, g	$T_{ign} = \frac{4190}{\log (A/m) - 5.23}$	$T_{ign} = 1343 - 208 \log (A/m)$
	Isothermal Oxidation Characteristics	
Nature of oxide	Nonprotective	Very protective
Diffusing species	Oxygen	Oxygen
Type of rate law	Linear, two stages	Cubic
Temperature range studied	125–295°C	400–900°C
Pressure dependence	Yes	No
Activation energy	Both stages pressure dependent, 12.4 to 20.5 kcal/mole	42.7 kcal/mole
Additive effects	Only very slight at 200°C[g]	At 700°C,[j] varied changes in rate law,[k] and breakaway phenomena[l]
Correlation to ignition data	None evident at 200°C	Weight gain in first minute at 700°C is inversely proportional to ignition temperature of 5-mil foil[i]

[a]These data are based on information obtained by Schnizlein et al. and reported in reference 44.

[b]Sample was heated at programed rate of 10 deg/min in flowing air (2500 cm/min) or oxygen (500 cm/min). Ignition temperature was determined graphically from time-temperature trace.

[c]Sample heated in protective helium to selected temperature before air or oxygen was admitted. If selected temperature was above ignition temperature, ignition occurred, but, if below, only mild self-heating occurred.

[d]Data obtained from ignitions in air were less distinct and more difficult to interpret because of tendency to thermocycle at lower temperatures (maximum burning temperature approximately 1500°C) than in oxygen.

[e]Ignitions in oxygen were spectacular and involved self-heating of bulk samples to temperatures greater than 2100°C.

[f]Data obtained in oxygen (see d and e above).

[g]Uranium additives studied: 0.5, 1, and 2 at.% Al, Be, C, Cu, Fe, Mo, Nb, Ru, Si, and Zr.

[h]Uranium foils studied: uranium A, uranium B, 1.0 at.% Al, and 1.0 at.% Cu.

[i]Zirconium foils studied: pure zirconium, 0.91 at.% Ni, 3.6 at.% Cu, 1.8 at.% U, 1.8 at.% V, 3.6 at.% Sn, 2.2 and 3.6 at.% Al, and 1.1, 2.1, 4.2, and 14.9 at.% Ti.

[j]Zirconium additives studied: 1, 2, and 4 at.% Al, Be, C, Cr, Co, Cu, Fe, Hf, Mo, Ni, Nb, Pb, Pt, Si, Sn, Ta, Ti, U, V, and W.

[k]Additives soluble in alpha zirconium at 700°C explained by Wagner-Hauffe theory.

[l]Breakaway phenomena observed for alloys in which additive ionic radius is more than 15 per cent different from zirconium ionic radius.

Fission-Gas Release from Reactor Fuels

Studies in the various fields of reactor development pertaining to the rate and extent of fission-product gas release may have as their basis either a fairly optimized system of fuel and its environment or, in contrast, a simulated fuel-element catastrophe ranging from simple melting in an inert atmosphere to the most serious situations, including a nuclear excursion and an autocatalytic metal-water reaction following a loss-of-coolant accident. These two approaches to fission-gas release investigations anticipate distinctly different results that must be determined from different experiments. It is also apparent that fission-product release during operation will occur under well-defined conditions of temperature, time, and environment, whereas the precise conditions to which the fuel element would be subjected in a catastrophe are generally a matter of conjecture. Knowledge of the upper limit on the activity that could be released during postulated catastrophes is of value, however, particularly in planning for and evaluating the consequences of such incidents. A previous article[45] in the December 1959 issue of *Nuclear Safety* discussed the data available at that time on fission-product release during various accident conditions. A review of recent literature on the release of fission products from UO_2 and from metallic uranium is given here.

Fission-Gas Release from UO_2

Diffusion studies of the release of rare gases from UO_2 heated in highly purified inert gas have been especially successful. As indicated in a previous article[46] in the September 1959 issue of *Nuclear Safety*, the application of simple diffusion theory to the release of fission products from UO_2 has been supported by much experimental evidence, although many of the factors influencing gas release are not clearly understood. More recent experiments have demonstrated the importance of the oxygen-to-uranium ratio and the effect of grain growth on the rate of release.

The significance of the bulk density of sintered or compacted UO_2 with regard to fission-gas release, as well as the closely related oxygen-to-uranium ratio parameter, as reported by Belle[47] and by Lindner and Matzke,[48] resulted in further experiments by Auskern[49] involving sintering anneals of irradiated UO_2 powders. The effect of the continuously changing particle radius as a result of grain growth at high temperature was demonstrated in Auskern's experiments, and the release values were used in calculating the diffusion coefficient, D, for the particular particle radii involved. The particle sizes of two samples at 1000°C were 0.22 and 2.8 μ and gave D values of 1.3×10^{-15} and 4.3×10^{-17}, respectively. After sintering at 1400°C, the radii had increased to 1 and 14 μ, respectively, and the corresponding D values had changed to 5.9×10^{-13} and 1.0×10^{-13}. The release of Kr^{85} at 900°C before sintering of the 2.8-μ powder amounted to 1 per cent in 5 hr. Auskern estimated that if the material had been previously sintered to the 14-μ diameter, the release at 900°C would have been only 0.04 per cent. These D values for sintered UO_2, when plotted against the reciprocal of the temperature, give an Arrhenius activation energy with values ranging from 46 to 80 kcal/mole in the temperature range of 1000 to 1500°C.

The superiority of stoichiometric UO_2 (oxygen-to-uranium ratio of 2.00 to 2.02) over nonstoichiometric UO_2 (oxygen-to-uranium ratio of 2.02 to 2.14) with respect to gas retention has been clearly indicated by Morgan et al.[50] Release values were determined after irradiation, for periods of one to six months, of various grades of sintered UO_2 with predetermined oxygen-to-uranium ratios. The maximum estimated burnup exceeded 20,000 Mwd/metric ton, and the central UO_2 temperatures ranged up to 3100°F. Nonstoichiometric UO_2 gave high fission-gas release values (2 per cent) at central temperatures as low as 2200°F, whereas the stoichiometric material approached a central temperature of 2700°F before giving the same release. Representative data obtained in these studies are shown in Table II-2.

It should be noted that D values calculated from these data are distinctly lower than those of Belle[47] or Auskern.[49] The data of Morgan et al.,[50] although extremely favorable for stoichiometric UO_2, show that above 2700°F there is a rapid increase in gas release. This may indicate phase transformation as a result of grain growth or uranium vaporization.

Heating sintered UO_2 in a corrosive or oxidizing atmosphere is known to result in serious physical and chemical changes; how-

Table II-2 FISSION-GAS RELEASE FROM UO_2

Capsule No.	Bulk density, % of theoretical	Oxygen-to-uranium ratio	Carbon content, ppm	Central UO_2 temp., °F	Estimated burnup, Mwd/metric ton	Gas release, %	
						Kr^{85}	Xe^{133}
L-6a	73.63	2.02		2100	14,000	1.9	0.3
L-2b	84.4	2.129	2500	2200	3,600	13	9.0
L-11b	85.23	2.14	630	2400	22,000	40	5.3
L-93b	74.87	2.118	2100	3000	4,600	24	19
L-25b	94.90	2.02	200	2500	5,100	0.15	0.22
L-16xa	93.98	2.005	70	2700	3,200	1.9	
L-16xb	94.35	2.005	70	3000	3,200	6.4	
L-17xb	94.83	2.005	70	3100	3,100	24	61

ever, recent melting experiments conducted in air[51] by the arc-image-furnace method have produced the first direct information on the nearly quantitative release of the rare gases, iodine, tellurium, ruthenium, and cesium. The simultaneous observation of vaporized uranium oxide led to confirmation of its composition as UO_3 in the vapor phase. For short heating periods of about 1 min, the release of the rare gases is apparently not complete (~75 per cent) and is sometimes exceeded by the release of iodine or tellurium (~80 per cent).

The excessive pressures (up to 60 atm after extensive burnup) calculated for a high-temperature nuclear excursion in which the fuel temperature ranges[52] from 2500 to 4500°C in a reactor fueled with graphite-clad graphite-impregnated slugs have led to the expectation of increased reactivity due to the loss of Xe^{135} and Sm^{149} from the core. The further possibility exists that I^{135} would diffuse out of the fuel slug under normal operating conditions, and therefore all the Xe^{135} could be released if the cladding were to break. Melting of carbide particles in the fuel would allow for diffusion of iodine and samarium vapor. Both these elements would therefore be found outside the fuel particles. The danger would be minimized by the use of relatively impervious graphite in the fuel element.

Fission-Product Release from Metallic Uranium

Melting and burning experiments have been carried out on metallic uranium in air,[53,54] steam,[53,55] CO_2,[53] helium,[53] and mixtures of these gases. The oxidation that occurs simultaneously with each of the heating operations leads to almost uniformly increasing amounts of total fission products released. Melting and burning in air are most disastrous, especially at the higher temperatures, and result in quantitative release of xenon and krypton (except at trace level), as well as most of the halogens and tellurium, some of the cesium and ruthenium, and less than 0.2 per cent of the strontium. When the burning was carried out in a carbon dioxide atmosphere, much lower amounts of the nonvolatile elements were released, but the rare-gas release rate was essentially unchanged.

In steam, a tightly bound UO_2 layer was surprisingly effective in retaining most of the rare gases (~3 to 5 per cent released at 1200°C in 2 hr), although at 1400°C some changes apparently occurred in the oxide layer and resulted in about 50 per cent release. In a 1200°C helium atmosphere, which probably contained traces of oxygen, the rare gases and halogens were released to an extent that was only slightly less than the release in air at the same temperature. For trace levels of fuel burnup, there was definite evidence of about a factor of 10 suppression of the release of the rare gases, cesium, and strontium when compared with release rates for 0.2 at.% burnup.

These experimental data on fission-product release may be used to predict activity release under the accident conditions which the experiments simulate. An indication of utility of the experimental data for such predictions may be realized by a comparison of the results of the analysis of the Windscale release with the experimentally determined ratios of the various fission products released from metallic uranium. Such a comparison is presented in Table II-3. The calculated release percentages for the several isotopes were derived from meas-

Table II-3 ANALYSIS OF WINDSCALE FISSION-PRODUCT RELEASE

Nuclide observed	Activity released* relative to I^{131}	Activity in fuel† 4 days after shutdown relative to I^{131}	Calculated activity release,‡ %	Predicted activity released,§ %
I^{131}	1	1.0	70	70
Te^{132}	0.58	0.75¶	54	70
Cs^{137}	0.03	0.08	23	18
Sr^{89}	0.01	1.6	0.44	<0.2
Ru^{103}	0.02	1.0	1.4	1.7
Zr^{95}	0.001	2.2	0.03	0.04

* Based on fallout filter samples; data from Booker[56] and Chamberlain.[57]

† Based on irradiation for 360 days.

‡ Based on Windscale data.

§ Based on experimental data described in this review.

¶ Ratio includes decay from time of reactor shutdown.

ured values of fallout on filter samples. The calculated value was multiplied in each case by a factor of 0.7, which is the known I^{131} release fraction from a sample that is 85 per cent oxidized. The predicted release percentages are based on experimental data at 1200°C, which is to be compared with the maximum temperature of 1300°C reported in the Windscale incident. The predicted values for release of fission products are in rather good agreement with the observed values.

(G. W. Parker)

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Section III

INSTRUMENTATION AND CONTROLS

Monitoring of Gaseous Wastes

The used air from a chemical plant is normally polluted with dust, noxious vapors, and various gases, and it may constitute a hazard to plant personnel, as well as to the general public in the environs of the plant. In many ways, the problems are similar to those which arise in dealing with the hazards of atomic-plant waste water.[1] These concentrations,[2] listed in units of microcuries per cubic centimeter, require that a representative proportional sample be used for monitoring purposes and therefore that the total gas flow be known. Other similarities in the problems associated with liquid and gaseous wastes are the necessity for maintaining a continuous record of radioactivity released to the public domain and the fact that certain isotopes generally emerge as the limiting or most hazardous ones at a particular operation. The logical point for sampling both types of waste is where control of the waste is relinquished.

There are, however, some obvious differences in handling liquid and gaseous wastes. The physical state of a liquid allows it to be held in large quantities in inexpensive ponds for days, and therefore ample time is available for processing. Furthermore, several recycles can be used if one processing cycle is not sufficient. In contrast, the large quantities of air (typically 50,000 to 100,000 cu ft/min) that pass through a plant must be cleaned instantaneously because the holdup of such quantities of gas is impractical in most cases. Also the MPC's for gaseous wastes are about 1/10,000 of the MPC's for water. In general, the liquid state is less difficult to handle than the gaseous state.

As a reference point for this discussion, a radiochemical plant that disposes of low-activity-level gases through a stack disposal system is assumed. The disposal problems in reactor plants from which large quantities of low-level activity would be discharged would be similar, but the types of activity might differ significantly.

In a radiochemical plant, air flows through the plant, and, in passing through processing cells to cool equipment containing radioactive materials, it becomes slightly radioactive itself. The activity in radiochemical plants is mainly particulate and is removed by high-efficiency filters before the gas is discharged into the disposal stack. A lower flow vacuum system is usually employed to collect the higher level off-gas from process vessels and pass it through scrubbers for removal of the volatile components and filters for the removal of particulate matter. Another method of cleaning is to trap volatile materials, such as iodine, in charcoal or metallic filters.[3–5] Rare gases cannot be easily removed although charcoal traps can delay them for appreciable decay times. Even though the cleaners are very efficient, they are not perfect, and therefore particulate matter, rare gases, and active gases may be expected in the stack gas when it is discharged to the atmosphere. Although additional dilution and dispersion occur after the gas has left the stack, control is lost when it leaves the stack. The gas may be monitored as it leaves the stack, although complete evaluation of the adequacy of the disposal system would involve spot checks at points in the environs of the plant up to several miles.[6–9]

Monitoring air for radioactivity is difficult because of the background contamination buildup in effluent monitors and because the MPC's for

most isotopes are minute. In order to achieve the recommended MPC levels, it is necessary to clean the air to such great purity that the concentration of the radioactivity is so low as to make it very difficult to detect and measure. The remaining activity must be reconcentrated by air-cleaning methods to get a reliable count; thus air monitoring and air cleaning become interdependent.

Obviously a stack monitor cannot be a single instrument, but, rather, is a system that employs several instruments, together with the possible refinement of a data-reduction block to make corrections and to provide a read-out of calculated data. Many stack monitors are in existence, but not much literature on specific systems has been published. Many stack monitors have been supplied by commercial instrument manufacturers, and the literature describes primarily the component parts rather than the total monitoring system. The component parts of the stack monitor are derived from laboratory air monitors since the same design concepts are usually involved.[10]

Evidence has been presented that the possibility of ingestion of particulate matter is the worst hazard[11] of air contamination, and the greatest effort in the development of air monitors has been on particulate-matter monitors that use filter paper as a collection medium.[9,12] The problem is complicated by the effects of particle size. Retention of solid matter by the lung is a function of particle size,[13] and samplers and monitors must be designed so that critical particle sizes are not discriminated against. A ramification of the problem is the necessity of evaluating the collection efficiency of filters and filter papers.[14] Other methods of collection, such as precipitation[15] and impaction, have been used. The impaction method allows actual separation of particles by size and has been utilized to reduce alpha background by discriminating against radon-bearing particulates in an alpha monitor.[16] Filter-paper samples can be monitored continuously or intermittently for beta, gamma, and alpha activity, or they can be taken to the laboratory for gamma-ray spectrometry. Instruments are commercially available which use a filter tape that moves under a beta-gamma detector and then, after a delay to allow radon-thoron background to die out, under an alpha counter. Essentially instantaneous alpha information can be obtained by subtracting the background[15,17] or by determining pseudocoincidences with beta-gamma emissions.[18]

The gaseous wastes from fission, such as I^{131}, Ru^{103}, and Ru^{106}, are difficult to collect and measure because of their reactive properties. Collecting a gas sample from the stack and transporting it to the instrument for measurement usually result in a loss of material since such materials react with, and plate out on, metal surfaces. They may also quickly become particulate by settling on dust particles or by changing state, and therefore separation of a sample into particulate and gaseous components to evaluate the activity level is ambiguous. Iodine will collect poorly on a filter; but, if total I^{131}, for example, must be known, considerable care must be used in adding the material lost to walls and filters before the iodine is trapped as such. Iodine is frequently trapped on charcoal, but, where rare-gas activity is present, some way must be found to separate the counts from this component because the rare gases are also trapped in charcoal.

The active gases, of which I^{131} is the most common offender, are monitored by collection in a charcoal trap, which, in turn, is monitored by a scintillation counter[19] or a Geiger-Mueller tube detector. An alternate method is to use a small scrubber to collect the iodine and then to monitor the scrubber solution. Hanford increases the accuracy of the determination by adding a computer to a system consisting of a filter and a scrubber so that the iodine contributions to each collection method are found and added correctly.[20] In still another method[21] used at Hanford, the radioactive gas is passed through a chamber which is monitored by a scintillation counter and arranged for chemical cleaning as the background builds up on the walls of the chamber. The quoted sensitivity of this monitor is 0.0725 μc/cu ft. A scheme[21] for correcting the readings of gross activity for the effect of Kr^{85} and Ru^{106}, both of which may be present with the iodine, has been described in Report HW-38483.

After all other components have been collected, the rare gases will generally remain in the air stream. Total activity can be gauged by passing the gas through or around an ion chamber, although collection in a charcoal trap at a low temperature[22] (−78°C) gives a more precise determination. At lower temperatures the efficiency of charcoal for the retention of the rare gases increases, and holdup can be accomplished

for long periods. By heating the trap, the collected sample can be transferred to a counting chamber for measurement of the activity level.

An important feature of stack monitoring is the alarm and catastrophe function of the monitor. The determination of the activity ranges to be covered by any given part of a stack monitoring system is mainly a matter of experience, and an unpredicted accident might throw readouts off scale so that for record purposes no instrumentation would be operating for the duration of the burst. In such a case an automatic device for taking a grab sample of the effluent air for laboratory analysis should be provided to fill a gap in the information necessary to evaluate the hazard of the burst. Sampling techniques are outside the scope of this discussion, but they are a most important feature of any stack monitor. A reasonably large amount of literature exists on this subject and on methods of testing sampling systems.[23] (D. J. Knowles)

Predicting Reliability of Safety Devices

Statistical analysis can be a valuable tool in predicting the reliability of reactor safety devices. A considerable amount of work has been done in this field, and three recent reports are reviewed here.

A paper by Connor[24] of The Research Triangle Institute illustrates how the failure data for a device can be fitted with probability distributions quite successfully if the data are first sorted into sets according to the time during which the failure occurred. Connor begins by showing two frequency distributions of unsorted failures of an unnamed device. One is a set of failures that occurred during the month of manufacture, and the other is a set of failures that occurred four months after manufacture. To these data he has fitted both the Poisson probability distribution and the negative binomial probability distribution. The Poisson distribution is given by the expression

$$\frac{e^{-m}m^i}{i!} \qquad i = 0, 1, \ldots$$

and the negative binomial distribution by

$$\binom{r+i-1}{i} p^r(1-p)^i \qquad i = 0, 1, \ldots$$

in which i represents the number of failures, m is the probable number of failures, r is a positive integer, and $0 < p < 1$. In both cases the Poisson fit, when tested by the χ^2 goodness-of-fit test, was judged to be poor; whereas the negative binomial fit was judged to be acceptable. Connor points out, however, that the negative binomial distribution cannot be used to describe the manner in which failure distributions change with time.

The failure data were then separated into several sets, each set being comprised of the failures that occurred during a given month. To the set of data which was classified as initial failures, Connor fitted the geometric probability distribution, which is given by

$$(1-w)^i w \qquad i = 0, 1, \ldots$$

where w is defined as the probability that a device passes every test in a battery of tests. The agreement of the fitted geometric distribution with the observed distribution was very good. The other distributions, for sets of data representing failures in subsequent months, were fitted with the Poisson probability distribution. This time the Poisson probability was determined from the expression

$$\frac{e^{-\lambda t}(\lambda t)^i}{i!} \qquad i = 0, 1, \ldots$$

where λt takes the place of m in the earlier Poisson formula. The constant λ is the average number of failures per unit of time (in this case, one month), and t is a time constant representing the number of months from manufacture to failure. The parameter λ is estimated from several months of data, not all of which are given in the article, by dividing the total number of failures experienced by the total number of unit months.

Connor then gives two failure distributions to which the Poisson distribution has been fitted using $t = 2$ and $t = 4$. The agreement of the Poisson fit with the observed data is acceptable. Also given are Poisson fits in which each of the sets of data is treated as a separate set of failure data. As might be expected, these latter calculations show better fits than the jointly fitted Poisson. A point is made that, with the parameter λ determined, future failures can be predicted by an appropriate selection of t. For example, the failure distribution 12 months after manufacture would be predicted by setting $t = 12$.

There is uncertainty in this prediction, of course, and Connor points out that the uncertainty would grow as the extrapolation is carried further into the future.

Connor concludes his paper with the presentation of a failure probability model for a failure occurring during the time interval $(t, t + dt)$. The model is

$$\lambda_n(t) = c\,\frac{1 + an}{1 + bt}\,dt$$

where a, b, and c are nonnegative constants, n is the number of previous failures, and t is the time since the unit was manufactured. It is stated that, if $a = b = 0$, the model reduces to that of the Poisson process. If $a \neq 0$, then, for a fixed t, the probability that a unit may fail increases with the number of past failures; this increase could occur if, for example, repairing a unit degraded it. If $b \neq 0$, then, for fixed n, the probability of failure would decline with time.

An application of sequential analysis to the determination of reliability of a double-diaphragm safety device for use in swimming-pool or tank type reactors has been described by Fitch.[25] An elementary exposition of sequential analysis and a description of the double-diaphragm device, together with a proposal for estimating the reliability of the device by sequential methods, are also presented.

Probably as a result of an effort to make the statistics simple, some inadequacies exist in the description of the statistical method. There is the implication that a normal sampling distribution is dependent upon all samples being of a fixed size. This is not necessary. It is also implied that, regardless of the sampling procedure used, the mean of the sampling distribution will be the same as the mean of the population. This also is not true. Furthermore, in developing formulas for the sequential method of analysis, Fitch indicates that a transformation to natural logarithms is necessary; whereas a transformation to logarithms on any base would suffice. Fitch also fails to mention that the formulas are approximations and that the size of the rejection risk, α, and the size of the acceptance risk, β, determine how good these approximations are.

The test procedure described allows the reliability of the safety device to be estimated at 95 per cent or more or at 85 per cent or less. The probability of declaring the device at least 95 per cent reliable when it is less than 95 per cent reliable is 5 per cent. The probability of declaring the safety device to be not more than 85 per cent reliable when actually it is greater than 85 per cent reliable is also 5 per cent.

For the tests, 30 safety devices will be manufactured, and, after accumulating shelf-lifetime or environmental-lifetime information, they will be shipped to the SPERT-I reactor for transient tests. A fuel element will be removed from the SPERT-I core and replaced with the safety device. A power excursion will be initiated, and an observation will be made as to whether the safety device passed or failed. The devices will be tested one at a time, and, after m such tests, d_m, the number of failures, and g_m, the number of successes, will be reported ($m = 1, 2, \ldots$). At the end of each test, the following decision rule will be used.

Accept R_1 if

$$d_m \log \frac{1 - R_1}{1 - R_0} + g_m \log \frac{R_1}{R_0} \geq \log \frac{1 - \beta}{\alpha}$$

Test another device if

$$\log \frac{\beta}{1 - \alpha} < d_m \log \frac{1 - R_1}{1 - R_0} + g_m \log \frac{R_1}{R_0} < \log \frac{1 - \beta}{\alpha}$$

Accept R_0 if

$$\log \frac{\beta}{1 - \alpha} \geq d_m \log \frac{1 - R_1}{1 - R_0} + g_m \log \frac{R_1}{R_0}$$

where α = rejection risk = 5 per cent
β = acceptance risk = 5 per cent
R_0 = desired reliability = 95 per cent
R_1 = alternative reliability = 85 per cent

Fitch has calculated that, with these test parameters and with this decision rule, the average sample number (the number of tests necessary to reach a decision on the average) will not exceed 30. The average sample number is an *average* sample size, however, and no rules are given to follow in the event that all 30 devices have been tested before reliability R_0 or reliability R_1 is accepted.

Calvert[26] of the Sandia Corporation has considered a situation in which a binomial variable (i.e., a variable that is classified as either a

success or failure after a trial) is drawn at random from a population of lots whose parameters follow the beta probability distribution. This reviewer has misgivings about the applicability of this theory since the assumption of a beta probability distribution for the parameters would appear to be quite arbitrary.

In Calvert's report[26] lot L is said to contain a fraction f of successful components. The binomial parameter, f, is assumed to have a probability distribution, $g(f)$, which is a beta distribution with parameters r and s. Calvert shows how confidence intervals on f can be constructed from the results of testing a number of components drawn at random from the population of lots.

Included in the report is a table of 95 per cent one-sided confidence limits for samples of sizes 10, 20, 30, 40, 50, and 60 with 0, 1, 2, or 3 failures observed when the samples are drawn (1) from populations in which nothing is known about the distribution of f, (2) from populations in which f is distributed as a beta variate with parameters $r = 24$ and $s = 1$, and (3) from populations in which the beta parameters are $r = 48$ and $s = 1$. Also diagrams of the beta distributions for $r = 24$ and 48 and $s = 0$, 1, and 2 are given.

For a one-sided, $1 - \alpha$, confidence interval of the form $(h,1)$, the formula for determining h is

$$\alpha = 1 - \sum_{i=a+r+1}^{a+b+r+s+1} \binom{a+b+r+s+1}{i} \times h^i (1-h)^{a+b+r+s+1-i}$$

where r and s are as defined above, a is the number of successful components, and b is the number of unsuccessful components observed in sample of size $a + b$. The symbol h may be interpreted as a measure of reliability of the components in that one might say, with $1 - \alpha$ confidence, that the reliability of the component is h or greater.

In general, the paper is well written, and the derivation of the formulas is clearly presented. However, the language is not clear at some points; for example, the formula

$$\int_0^1 f\, g(f)\, df = \bar{f}$$

is defined as the probability that a unit drawn at random from lot L is successful. The integrand would be properly defined that way; however, the integral defines the probability that a unit drawn at random from the population of lots is successful. In another case, the author computes a probability of observing three or more failures in 10 trials as 0.056 (the reviewer obtained 0.057) and states that a change in the manufacturing or the inspection process would not be suspected on the 0.01 level but probably would be on the 0.05 level. Since 0.056 is greater than 0.05, a change should not be suspected at the 0.05 level either.

(Donald A. Gardiner)

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Section IV

PLANT SAFETY FEATURES

Penetration of Reactor Containment Shells

By DONALD E. DAVENPORT*

Reactor containment is usually the final safety device in the complex safety system of any reactor. The complexity of the entire safety system and the effort and money invested in assuring safety testify to the importance of safety in a reactor. Although it is generally agreed that the probability of a major reactor accident is very low, as indicated by any method of calculation, the potential damage[1] from the release to the public domain of a significant fraction of the fission products in an operating power reactor makes such a release "unthinkable."

Compared with many industrial explosions, the energy released in a major reactor accident would be quite modest. Even under extreme conditions, the postulated reactor accident would yield less energy than that of 1 ton of TNT, which is to be compared with 1000 tons at Texas City in 1947, or 6 tons at the Whiting Refinery in 1955, or 6 tons in the explosives truck fire at Roseburg, Oreg., in 1959. The release of all the radioactive fission products stored in the fuel of the reactor could, however, force evacuation of large areas and cause severe illness or death to many of the inhabitants of nearby areas.

Because reactor containment is the "last-ditch" safety device, it is particularly important that it really offer full protection in case of any credible accident. Since it encompasses the entire system, it is large in size and expensive to construct. Therefore its cost becomes an important item in the total cost of the electric power that the reactor will produce, and, as the over-all cost of nuclear electric power approaches that of conventional electric power, the cost of the total containment system becomes proportionately larger for a fixed cost of containment. For these reasons the outer containment of reactors has received a good deal of attention in terms of contract work initiated by the AEC and by power-reactor designers. Contract work on this problem is under way at Stanford Research Institute, Armour Research Foundation, Naval Ordnance Laboratory, Ballistic Research Laboratories, General Electric Company, Argonne National Laboratory, and Sargent & Lundy.

The various techniques of containment, i.e., steel pressure shells, vapor condensation, and vapor venting, have been discussed in previous

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issues of *Nuclear Safety*.[2–5] At the present stage of development of power reactors, it is particularly important that all containment concepts be evaluated thoroughly, first as to their contribution to safety and then as to their cost. In this discussion, only one phase of the protection aspect will be examined, with particular emphasis on the threat of missiles to the containment system.

The containment concept was first regarded as the "brute-force" method which would offer full protection in any event. It was soon realized, however, that the integrity of the shell might be threatened by a reactor incident, even though the shell was built to withstand the anticipated overpressure. The additional hazard stemmed from the missiles that might be generated in a reactor runaway; for example, a displaced step-plug, an ejected control rod, or even large chunks of the concrete biological shielding. In the event of a pipe rupture or the failure of a vessel closure, large pieces of steel pipe or plate might become missiles. It is clear from such examples that, in order to evaluate the safety of a nuclear power plant, consideration must be given to the kinds of missiles that can threaten the integrity of the containment system and to the conditions under which such missiles can be generated. Hopefully, the study of the problem will lead to methods of protecting against or preventing the generation of such missiles.

The Hazardous Missile

In evaluating the hazard to the simple steel vapor container, use may be made of penetration data obtained at the Stanford Research Institute* from studies of steel plate. The data indicate that the minimum energy (E) per unit diameter (D) of missile, in foot-pounds per inch, needed for penetration of a steel plate is given by the equation

$$\frac{E}{D} = U(0.34T^2 + 0.032T) \qquad (1)$$

where U is the ultimate tensile strength of the plate, in pounds per square inch, and T is the plate thickness, in inches. Since Eq. 1 is valid only for missiles that do not increase in diameter during penetration, the results may be regarded as conservative when applied to missile materials softer than steel. The equation is based on a rigidly held plate, and it predicts somewhat lower energies for penetration than would be required if the boundaries of the plate were distant from the point of impact, as would be the case for most areas of the vapor shield. Such a conservative equation is, however, desirable.

On the basis of Eq. 1, the most hazardous missile for given mass and velocity is rod shaped and, in its most hazardous impact, strikes endwise, since this gives the highest E/D. If the projectile has a conical tip, the diameter of interest is that at which penetration is just achieved (i.e., the diameter 1 in. from the tip for 1-in.-thick vapor container), since penetration to this depth will make a hole in the container, even if the missile does not go completely through.

A useful form of the term E/D is

$$\frac{E}{D} = \frac{mV^2}{4R} \qquad (2)$$

where m is the mass of missile, V is its penetration velocity, and R is the radius of the impact area at penetration. For a cylindrical missile striking endwise, Eq. 2 reduces to

$$\frac{E}{D} = \frac{\pi\rho V^2 RL}{4a} \qquad (3)$$

where ρ is the density of the cylinder, a is the gravitational constant, and L is the length of the cylinder having the radius R and the volume of the missile. For a given thickness of target and density of missile, the velocity of the missile may be plotted against the product RL as in Fig. 1.

If Eq. 2 is stated in terms of weight (W) of the missile, i.e.,

$$\frac{E}{D} = \frac{WV^2}{4Ra} \qquad (4)$$

the single curve that represents the penetration velocities of all missiles as a function of the missile W/R ratio may be plotted, as shown in Fig. 2. From curves such as those of Figs. 1 and 2, estimates can be made for the minimum velocities at which missiles can penetrate the vapor container. For example, the velocity required for penetration by a 4-in.-diameter steel

*Under AEC contract AT(04-3)-115.

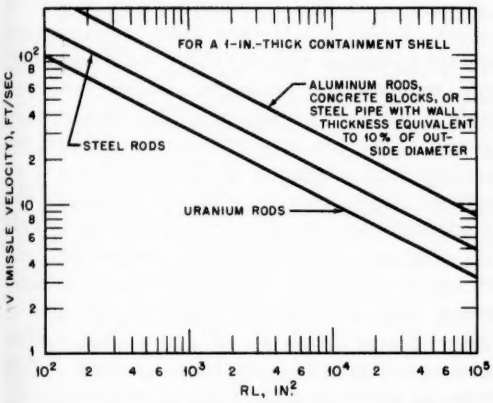
FOR A 1-IN.-THICK CONTAINMENT SHELL
ALUMINUM RODS, CONCRETE BLOCKS, OR STEEL PIPE WITH WALL THICKNESS EQUIVALENT TO 10% OF OUTSIDE DIAMETER
STEEL RODS
URANIUM RODS
V (MISSLE VELOCITY), FT/SEC
RL, IN.²

Figure 1—Penetration velocity as a function of missile RL product. (R = radius of impact area at penetration, and L = length of the cylinder having the radius R and the volume of the missile.)

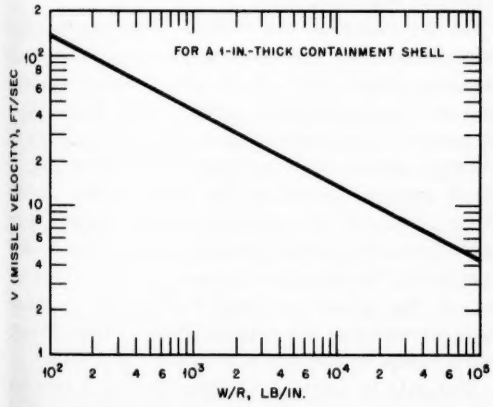

Figure 2—Penetration velocities as a function of missile W/R ratio. (W = weight of missile, and R = radius of impact area of penetration.)

rod 10 ft long, i.e., for $RL = 2 \times 120 = 240$ sq in., may be seen from Fig. 1 to be about 100 ft/sec.

The estimate may be further improved by considering for each material the minimum radius of impact area which will allow penetration without exceeding the compressive strength of the missile. Measurements have shown that, for a 1-in.-thick plate, the minimum radius of impact for penetration is at least 1 ft for concrete but is less than 1 in. for hardened steels. Thus, from Fig. 2, it may be seen that a 5-ton piece of steel traveling at 15 ft/sec could penetrate the 1-in. vapor container if the radius of the impact area were less than 1 in., whereas a block of concrete traveling at 15 ft/sec would have to weigh close to 60 tons in order to penetrate because concrete requires a radius of impact of 12 in. As the velocity of the missile increases, the minimum weight rapidly decreases; therefore, for a missile traveling at 50 ft/sec, the corresponding steel and concrete missiles would have to weigh 800 and 9600 lb, respectively.

Penetration of the vapor container requires velocities of over 100 ft/sec for missiles weighing less than 100 lb. Such velocities can be generated from the rupture of high-pressure coolant loops, but they appear to be unlikely from a reactor runaway unless the reactor vessel ruptures. From this point of view, the rupture of high-pressure coolant lines may constitute as great a hazard to containment integrity as that of a runaway reactor. In the case of a violent runaway, coolant-line ruptures will probably also occur when the shielding breaks up. If failure of the high-pressure loops initiates the incident, however, and there is no simultaneous or subsequent release of fission products from the melting of fuel elements, the importance of the integrity of the vapor container is very much reduced, since there would be no threat of a major radioactivity release.

Reactor Breakup During a Runaway

In order to determine the extent of breakup a reactor will suffer during a runaway and the nature of the missiles that can be generated, a series of small-scale (about 1:24) reactors are being built and tested by Stanford Research Institute using simulated energy sources in place of the reactor core.[6] It seems evident that at least four features of the energy release must be properly simulated if the models are to perform realistically: (1) the total energy release, (2) the rate of energy release as a function of time, (3) the partition of energy between solids, liquids, and gases, and (4) the geometry of the energy source. The energy sources that have been developed in this program are accurate with respect to total energy and have attained a good flexibility with respect to time-energy profiling, partition of energy, and geometry. As more is learned about the actual conditions in a reactor runaway, the energy sources can be made to include nonnuclear contributions such as the metal-water reactions, multiple bursts, and other complications. Present designs in-

corporate only the exponentially rising and decaying nuclear energy release. This simplified model of a nuclear burst allows the study of the importance of various reactor parameters on the damage produced.

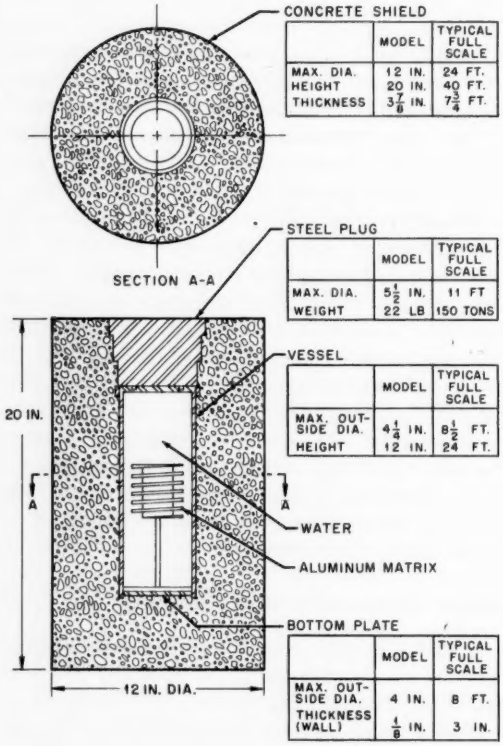

Figure 3—Model reactor for missile generation tests.

Casual consideration indicates that important reactor-design parameters which may affect the damage produced include (1) for the moderator, type, temperature, pressure, and percentage of voids in primary container; (2) for the primary reactor container, strength, volume of connected vessels, and presence of "blowout" ports or weak joints; (3) clearance between primary reactor container and shielding; (4) for the shielding, strength, shock-absorbing properties, and thickness; (5) for the primary container attachments, area of openings, mass and strength of closures, and flexibility of piping. The reference model reactor system being used in this study is shown in Fig. 3, along with model dimensions and typical full-scale dimensions. The model resembles, in general proportions, the typical pressurized-water and boiling-water reactor designs. The aluminum matrix in the center supports the energy source that simulates the reactor core.

On the basis of the first 70 reactor models tested, several interesting results have been obtained which will guide future experimental design. The results are presented in Table IV-1 in terms of equivalent total energy release in the typical full-scale reactor indicated in Fig. 3. Since energy scales as the cube of the linear dimensions of the system, these values are $(24)^3$ times the actual amounts used in the model reactors. The absolute values are characteristic of the particular materials used in these experiments and the original reactor conditions: room temperature and a pressure of 1 atm.

The data in Table IV-1 indicate that a reactor with its voids only 75 per cent filled with water requires about twice the energy release to give

Table IV-1 MINIMUM ENERGY REQUIRED TO FRACTURE CONCRETE SHIELD OF PRESSURIZED-WATER OR BOILING-WATER REACTOR BASED ON EXPERIMENTAL MODEL DATA SCALED UP BY FACTOR OF 24

Water content in reactor voids, %	Type of concrete	Air gap between reactor and shielding in full-scale reactor, in.	Reactor period,* msec	Energy in gaseous products, %	Minimum total energy required to fracture concrete shield, Mw-sec
100	Standard	0	1	20	250
75	Standard	0	1	20	550
100	Reinforced	0	1	20	750
100	Standard	6	1	20	1100
100	Standard	0	1	100	250
100	Standard	0	0.01	100	50
100	Standard	0	2	100	>400

*The duration of the runaway was simulated as 7 periods: $2\frac{1}{2}$ periods rising, 2 periods at peak release rate, and $2\frac{1}{2}$ periods falling exponentially back to zero.

the same damage as a reactor in which the voids are completely filled with water. The damage was estimated from the breakup of the outer shielding, as well as from the velocity with which the step-plug was ejected. The void space acts to relieve the shock wave more quickly and to lower the static pressure generated by the gas evolution. These results imply that the boiling-water reactor would be able to withstand more severe runaways, even at the short periods indicated in Table IV-1, than the pressurized-water reactor, which has no unfilled voids.

As would be expected, the data show that increasing the strength of the shielding, by the addition of reinforcing rods or by changing its composition, can increase its resistance to breakup. This does not affect the velocity with which the step-plug is ejected if the reactor does not rupture and release the high-pressure steam and water, since the plastically expanding primary vessel which drives the step-plug behaves in much the same way for either type of shielding.

The introduction of an air gap between the primary reactor vessel and the shielding markedly decreases the shock transmission and allows the primary vessel to absorb a larger fraction of the energy release in plastic flow. Table IV-1 shows that the air gap increases the minimum energy release for damage from 250 to 1100 Mw-sec. In a few experiments (not reported in Table IV-1) the closure was omitted from the primary reactor to simulate failure of the steel vessel at an early stage of the runaway. In these cases the step-plug served to confine the pressure within the concrete shielding until the concrete broke up, but the pressure was rapidly equalized inside and outside the steel reactor vessel. Under these conditions, little plastic flow of the vessel occurred, and the breakup of the concrete increased markedly over that obtained with a sealed primary container. Thus the energy absorbed by the primary container can significantly reduce the damage produced. These results are in agreement with the measurements made at the Naval Ordnance Laboratory,[7] which showed that strain-energy absorption is significant.

It is very interesting to note in Table IV-1 that the minimum energy release for damage does not seem to differ significantly between 100 per cent gaseous products and 20 per cent gaseous products. (To obtain only 20 per cent of the energy in gaseous products, pyrotechnic mixtures such as aluminum and barium peroxide are added to the explosive energy source to give solid reaction products.) This is not to say that the extent of damage produced if the energy is above this minimum is comparable, since the 100 per cent gaseous release is much more damaging. At 350 Mw-sec, in fact, the 100 per cent gaseous release appears to be as damaging as a 1400 Mw-sec release with 20 per cent gaseous products. Further tests will be made near the minimum energy point to verify the comparison. When there is an air gap between the primary reactor vessel and the shielding, there is reason to believe that the energy partition will be important even for minimum damage.

The duration of the energy release can be seen from Table IV-1 to have a marked effect on the minimum energy needed to produce damage. As the reactor period shortens, the total energy necessary to produce damage also decreases. This effect is the result of rate of energy release on the shock strength produced. For designs that use materials which attentuate the shock strength (such as the air gap or addition of shock-absorbing materials; for example, various types of wood[8]), this difference should be appreciably reduced. Future experiments will be designed to examine this effect.

Missile Generation

Examination of the results of simulated runaways involving more than the minimum energy to produce damage gives information regarding the size and velocity* of missiles produced in the runaway. In the models tested, the step-plug generally had a higher velocity than the large concrete fragments. For the 100 per cent filled models, the step-plug velocities varied from 10 to 20 ft/sec with energy releases between 600 and 2000 Mw-sec. For the 75 per cent filled models, the velocities were about half as great. Again the lowering of the peak pressures by the void has the expected effect in lowering the velocity of missiles. It should be emphasized that the results were for a reactor vessel which did not rupture, the acceleration of the missiles being produced only by the expansion of the primary reactor vessel. If the reactor vessel rup-

*Velocity is constant for scaled models since distance and time scale by the same factor. Thus the measured velocity for the small models would be the same as the expected velocity in the full-scale reactor.

tured, even for these cold-water experiments, the plug velocity was greatly increased.

The velocity of the concrete missiles varied from about one-tenth that of the step-plug for the largest pieces of concrete to equal velocity for the smaller pieces. Thus the pieces that scaled to 250- and 350-ton pieces had velocities of 2 to 4 ft/sec in this energy release range, whereas the 60-ton pieces may have had about twice that amount. Although such pieces would make impressive missiles if they reached the vapor containment shield, their low velocities assure that they will strike the floor of the reactor area before traveling very far. Table IV-2 gives the horizontal travel distance of a missile before striking the floor for starting positions at several heights above the floor.

Table IV-2 APPROXIMATE MISSILE TRAVEL DISTANCES BEFORE STRIKING FLOOR

Height of origin, ft	Horizontal travel (ft) of missile with velocity of 5 ft/sec	Horizontal travel (ft) of missile with velocity of 20 ft/sec
10	4	16
20	6	22
40	8	32
80	11	45

These results make it reasonable that reactor runaways which do not lead to the rupture of the primary reactor container or piping are not apt to generate missiles that will threaten containment even for fairly severe energy releases. For thinner concrete shields, shorter reactor periods, or more complicated hardware attached to the reactor, even this fairly mild conclusion does not apply, since missile velocities will be much greater. Future experiments should extend the range of measurements to include most designs of interest to power-reactor engineers.

For a simulated runaway of 3000 Mw-sec, the primary reactor vessel ruptured, and this rupture resulted in a plug velocity of about 100 ft/sec. The velocity of the concrete shielding, however, increased only the expected amount to 15 to 20 ft/sec. Since the step-plug was not rigidly held in place, it was accelerated early in the simulated runaway while the shielding was still intact and acted essentially as a gun tube. The loss of the step-plug allowed venting, which held the velocity of the shielding to reasonably low values. This emphasizes the effect that the rupture of the primary container will have on the velocity of some components even for a cold-water filling. If the water had been heated to 600°F, a much more spectacular increase would have been expected.

It seems clear that prevention of this kind of an incident with rupture of the primary container would do a great deal toward simplification of the missile hazard problem.

Conclusions

A modeling technique that can include most of the important features of a reactor and reactor runaway has been devised to make possible the detailed study of missile generation. The experiments to date have been carried out on simplified reactor models which have no piping connections, test holes, control rods, or other connecting hardware and which use cold water as a moderator. These experiments confirm the usefulness of the technique for qualitative evaluation of results and suggest that quantitative data could be obtained by including more of the important reactor features. Scaling of the results will be checked with larger models and by comparison of predictions with actual performance of reactors such as SPERT.

The experiments to date have demonstrated the behavior of the primary reactor vessel in a nuclear runaway. Since there can be no significant shock waves generated in the nuclear runaway because of the long energy release period—many milliseconds—the swelling of a pressure vessel is quite similar to blowing up a balloon. The elongation before rupture may be 15 to 30 per cent, depending on the amount of radiation damage, the rate of elongation, the details of construction, and other factors. In the case of a steel plug which touches the vessel, rapid swelling acts like a piston to drive the plug out of the reactor to appreciable heights even though the reactor does not rupture. In reactors where the concrete is cast next to the reactor vessel, the concrete may be similarly accelerated and broken for quite low energy releases. If a gap exists between the reactor vessel and the concrete, larger energy releases are necessary before the reactor vessel strikes the concrete and causes the breakup and acceleration phenomena to set in. In either case the net force acting on the concrete is increased if the vessel ruptures, since the restraining effect of the vessel is no longer present.

The following major factors have been demonstrated by the experiments to date:

1. The design of the primary container and the gap that separates it from the shielding have pronounced effects on missile generation by providing for large amounts of energy absorption in plastic flow of the primary container.
2. The damage to the concrete shielding is much less if the reactor vessel does not rupture.
3. The damage to the concrete shielding becomes greater as the period of the runaway becomes shorter and the fraction of energy going directly to gaseous products becomes larger.

(Donald E. Davenport)

Particulate-Filter Testing and Inspection Program

Since the inception of the atomic energy development program, the problem of control of radioactive particulate matter in waste gas streams has existed. In order to prevent possible area contamination, absolute filters have been used in off-gas lines for the capture of particles that might constitute a health hazard, but this method of control is admittedly less than perfect. The general public, as well as governmental bodies on all levels, has therefore shown ever-increasing concern about the amount and type of particulate matter being released to the atmosphere, particularly with regard to the possibility of the "fallout" type of area contamination with radioisotopes. The control of the air pollutants released by all industrial installations is of universal interest, and the control of atmospheric pollution by other than radioactive waste is regulated by local and state law; regulations implementing control of radioactive waste are, however, the responsibility of the AEC. A review of state laws clearly shows that the existing controls are inadequate in most localities for the control of radioactive waste pollution of the atmosphere.[9]

The requirement for effective air cleaning in the nuclear energy field, because of the expanding technology in this area, has created a demand for high-efficiency particulate removal under increasingly severe operating conditions. The dry type of air filter has long been used where high efficiency of collection has been required; however, high humidity, the presence of corrosive gases, and/or elevated temperatures have seriously limited the usefulness of this type of waste-gas control until recently. A wide selection of materials is now available for filter media and separators. These new materials provide for greater flexibility of application, and the previous limitations can be overcome at a reasonable cost.

The Oak Ridge National Laboratory (ORNL) has used a number of different types of absolute filter for the removal of radioactive particulate matter from waste gases. Only one type, in various sizes, has been made a standard stock item; others have been acquired for specific uses as the need has arisen. Because of the great increase in filter usage, it is planned to study current needs and those of the immediate future in order to ascertain what additional items must be stocked and in what quantities. To prepare for this, new and revised procurement specifications are being written, along with a detailed directory of information concerning each filter installation in the X-10 area. This directory is expected to supply maintenance personnel with the information needed for servicing each particular filter installation and for maintaining a service record for each.

The discussions last summer at the Sixth AEC Air Cleaning Conference held in Idaho Falls, Idaho,[10] concerning defective filter units being stocked by AEC contractors have created great concern. At ORNL, stocked filter units were given a close visual inspection. Many filters were found to be defective and were promptly removed from stock. The stock procured subsequently has also been found to contain defective units upon receipt from the manufacturer. One shipment of 50 units contained 6 units that were determined to be defective. This experience is not unique with ORNL. Other AEC installations have had rejection rates as high as 56 per cent on the basis of visual inspection of high-efficiency filters.[11]

The formation of filter inspection and testing services for AEC contractors at the Chemical Corps Arsenal, Edgewood, Md., and at Hanford, Wash.,[12] now provides means of at least ensuring contract performance in the procurement of high-efficiency filter units by subjecting all units to a dioctyl phthalate (DOP) particle-penetration test.[13] Obviously, the past procurement method has lacked a means for determining compliance with contract terms relative to tested efficiencies. It is the intent at ORNL to require all filter units to be inspected and sub-

jected to the DOP test before being added to stock.

Although use of the testing service is not mandatory, the AEC recommends its use to the fullest extent possible. The delay it causes has, however, presented a problem because of depleted stocks at a time when the need is increasing. The first ORNL purchase tested at Edgewood consisted of 132 units and required a period of 30 days from receipt at the Edgewood Arsenal to final receipt at Oak Ridge of the tested units. Five units of this group failed to meet the testing and inspection requirements, and the testing revealed another condition that was injurious to filter efficiency. In the past, ORNL has been specifying "mineral fiber" gaskets attached to fire-resistant filters. An added test for compliance with this specification showed that such gaskets impaired the efficiency of the assembled unit. Not once in the history of previous purchases had a manufacturer indicated that this shortcoming was apparent as a result of his DOP testing. Neoprene gaskets are now being specified on fire-resistant filters for operating temperatures up to 250°F.

As further insurance against stocking filters that have been damaged by shipping and handling, all units are visually inspected both upon receipt from the Edgewood Arsenal and immediately prior to installation. These practices are to be continued until ample evidence exists that the new and improved packaging can be relied upon. Such inspections of the 127 filters from the above-mentioned 132 units which showed satisfactory filter efficiency in the Edgewood DOP test have revealed 15 units with visible defects, principally cracks or tears in the filter medium occurring typically, but not necessarily, at the glue-to-filter medium junction. Other defects detected by visual inspection have consisted of tears caused by spacers pushing through the filter medium and holes in the filter medium; and some filters had been crushed by external pressures. The tears observed were as much as $\frac{1}{8}$ in. in width and constituted serious impairment of filter integrity.

Careful handling at the time of installation is also an important phase in the filter-replacement cycle. All the previous effort and expense is of no avail if care is not exercised during installation. There is no practical means of field repairing a torn filter medium, and therefore any damaged filters are lost completely. Allowing a damaged unit to be installed is equivalent to having no filter at all.

In an effort to be assured of proper installation, ORNL requires that all critical filter installations be inspected by personnel familiar with the handling procedures for such operations. This service, as well as all other visual inspection, is provided by the Inspection Engineering Department.

The final and most positive proof of proper filter installation is a test in place after installation. Since only a small portion of the area of a filter medium is visible, defects of the type described above in the filter medium-to-glue junction could exist at points where their detection by visual inspection would be impossible. Also, many ORNL filter installations involve replacement by indirect or remote means, that is, "in-cell" installation or installation in in filter pits where group clamping is provided to perfect the sealing. Visual inspection is seriously hampered under these circumstances, and a test after installation remains the only reliable method of inspection. It is proposed to utilize a limited filtration efficiency test. A polydisperse DOP aerosol with a particle population containing individual drops of up to 2.5 μ in diameter but with an average drop size of $\frac{3}{4}\mu$ will be used; this aerosol will be produced by a generator developed by the Naval Research Laboratory.[14] Upstream and downstream samples of this aerosol will be tested with a forward scattering photometer that measures concentrations, and these concentrations can be converted into measures of filter efficiency. This device is not intended to replace the standard 0.3-μ DOP test, which is a specification requirement and is performed at Edgewood Arsenal. The purpose of the poly-disperse DOP test is to detect relatively gross (as compared with a particle diameter of 0.000012 in.) leakage caused by damage to the filter medium in handling or by inadequate gaskets rather than low filter efficiency resulting from faulty manufacturing methods. It is anticipated that the combination of the Edgewood Arsenal DOP test, careful visual inspection, and the limited "on-site" DOP test will provide the reasonable assurance of adequate, reliable particulate removal from waste gases which will preclude the possibility of area contamination under normal operating conditions. (W. W. Goshorn and A. B. Fuller)

High-Temperature Fission-Product Traps

Fission-product contamination of a reactor coolant constitutes a serious problem in radiobiological safety. This hazard is common to all reactor systems to varying degrees. In circulating-fuel reactors, fission products representative of almost the entire spectrum are circulated in the primary loop unless provisions can be incorporated to remove a fraction of the fission products from the coolant, either continuously or at frequent intervals. The problem is less acute but is present in fixed-fuel reactors. In gas-cooled reactors many fission products are mobile in the high-temperature operating regions and can enter the coolant stream by escaping from the matrix of the fuel elements through various mechanisms. Aside from contamination through direct leakage from faulty fuel elements, radioactivity can be induced in the coolant or in the circulating corrosion products in fixed-fuel liquid-cooled reactors.

Penalties that are consequences of contamination of the circulating stream include increased shielding, increased difficulties in maintenance of the system, and, in case of an accident, enhanced risk to the operating personnel, to the reactor site, and to the surroundings. Consequently devices are incorporated in all reactor systems to prevent, or at least control, the degree of contamination of the circulating stream. Many methods of radioactivity control have been considered, including complete containment of fission products by adequate cladding of fuel elements, chemical reactions to precipitate and render immobile the hazardous materials, electrochemical separations, solvent extraction, volatilization, filtration, and physical separation. This review is limited to "on-stream" methods of extracting radioactive materials at temperatures above 300°C. Part of the discussion will be concerned with proposed methods for controlling radioactivity buildup in advanced reactor concepts, and some emphasis will be placed on new and unproved systems that hold some reasonable promise of success.

Molten-Salt-Fueled Reactor

A feature of the molten-salt-fueled reactor is that it might prove to be an efficient thermal breeder when operating on the thorium cycle. A molten-salt-fueled thermal breeder would consist of a graphite-moderated two-region reactor fueled with a molten mixture of Li^7F, BeF_2, and UF_4 and blanketed with a mixture of Li^7F, BeF_2, and ThF_4.[15] The operating temperatures of the system would range from 450 to 750°C.

Xenon and krypton gases can be effectively removed from a circulating molten-salt fuel by a relatively simple method, which has been discussed in a number of articles.[16–19] The removal system being considered is based on the extremely low solubility of the noble gases in molten fluorides. In the present design a sufficiently large gas space is provided to permit the noble gases to come out of solution and to form a gas phase. Xenon solubility data[20–23] show that the ratio of the concentration of xenon in a gas phase to the concentration of xenon in solution in a molten fluoride-salt mixture is about 6000 at 600°C. Thus, increasing the volume of the gas phase in contact with the liquid results in a decrease in the amount of dissolved xenon in the salt. For example, utilization of an expansion tank with a volume equivalent to one-half the salt volume should decrease the xenon concentrations to $1/3000$ of its original value in the absence of diffusion into the pores of the graphite moderator. The corresponding reduction for krypton would be about $1/1500$. The xenon and krypton thus extracted from solution would be continuously removed from the expansion tank by a flushing stream of helium and transported to charcoal adsorbers where they would be "held up" to permit decay. Such a simple high-temperature fission-product trap is indeed attractive, even though the removal process is complicated by the beta decay heat that is generated in the expansion tank or in the proposed adsorbers, such as charcoal beds.

Recent calculations[18] of the quantities of fission gas that might be removed by such a procedure have suggested that the system may be very satisfactory. The characteristics of the reactor model chosen for the calculations are given below:

Power, Mw	500
Power density, kw/liter	230
Total fuel volume, cu ft	137
Average flux, neutrons/(cm²)(sec)	2.5×10^{14}
Total core volume, cu ft	480

Graphite volume, cu ft	403
Graphite void volume, cu ft	80.6
Fuel volume in core, cu ft	77
Expansion tank volume, cu ft	35

It was assumed that all the iodine and bromine would remain as soluble ionic halides in the liquid fuel and would not be adsorbed by the walls. The possibility of the graphite pores filling up with Xe^{135} was, however, considered. For the calculation, the diffusion coefficient of xenon in graphite was varied as one of the parameters, and the helium flushing rate of the expansion tank was varied as another parameter. Xenon poison fractions were calculated as functions of these parameters.

The results of the calculations indicate that most of the Xe^{135} can be removed by this method. For example, 98 per cent of the Xe^{135} formed could be removed by using a helium flushing rate of 2 cfm, based on 10^{-5} cm^2/sec as the diffusion coefficient of xenon in graphite. With the development of more nearly impervious graphite, a value of 10^{-5} cm^2/sec for the diffusion coefficient of xenon in graphite appears to be realistic. The cost for the noble-gas removal system, including decay tanks, charcoal beds, blowers, and necessary heat-dissipating devices, has been estimated to be 0.05 mill/kw-hr for the reactor model chosen.[18]

The possibility of good performance with this method of gas removal has supporting evidence from the operation of the Aircraft Reactor Experiment, in which the pump served as an effective degassing device, and only a very small fraction of the anticipated xenon poisoning was observed during operation, even though the system contained no special apparatus for xenon removal.[24] Moreover, many of the fission-product activities are noble gases for a period of time in their decay chains, and therefore the degree of contamination from the sources would be considerably reduced through effective gas removal by the method described.

The rare-earth elements account for a significant fraction of radioactive contamination of the circulating stream by nonvolatile fission products. These elements will be present as trivalent fluorides in the fuel of a molten-salt-fueled reactor. The possibility of removal of the rare-earth fission products from the circulating stream at high temperatures has been studied in some detail, and several methods have been discovered.[25–28] These methods include removal by exchange with cerium through formation of solid solutions with an excess of solid cerium fluoride, precipitation as oxide phases, and removal by isotopic exchange between dissolved radioactive solutes and nonradioactive insoluble solids. However, these methods qualify better as reprocessing schemes than as simple high-temperature fission-product traps, and, accordingly, they will not be discussed here.

Liquid-Metal-Fueled Reactor

The liquid-metal-fueled reactor concept envisions a two-region thermal breeder employing a fuel consisting of a dilute solution of enriched uranium, magnesium, and zirconium in liquid bismuth. The volatile fission products consist principally of krypton and xenon, with small amounts of bromine and iodine. Means for removal of these volatile fission products from the LMFR have been discussed in several reports.[18,29,30] In the absence of complications, the problem of noble-gas removal from the LMFR is quite similar to that for the molten-salt-fueled reactor. Experimental evidence of concentration of iodine at fuel-graphite interfaces and diffusion into the graphite[29] has, however, revealed a problem which requires further study. The diffusion and absorption of halogens and noble gases in graphite may be considerably lessened with the advent of less permeable graphite.

The continuous removal by high-temperature liquid-liquid extraction of some nonvolatile fission products with the use of a mixture of molten chloride salts has been successfully tested in small-scale experiments and has been discussed by Lane et al.[31] This removal method will not be discussed here because, as in the case of the molten-salt reactor, it is considered to be a reprocessing scheme rather than a simple high-temperature fission-product trap.

Aqueous Homogeneous Reactor

The biological hazards associated with a homogeneous reactor are due chiefly to the radioactive rare earths, alkaline earths, and iodine.[32] A major fraction of the radiation hazards is contributed by I^{131}. Detailed information on iodine-removal procedures for use with aque-

ous homogeneous reactors is available.[18,33–36] The use of silvered metallic beds to remove iodine vapor from the system is a method that is applicable at high temperatures, and the metallic beds may be classified as high-temperature fission-product traps.

Many of the fission and corrosion products in uranyl sulfate solutions of the type used to fuel the HRT* at temperatures in the range 250 to 325°C exhibit only limited solubility, with a negative temperature coefficient. Accordingly, their accumulation in the reactor system may be controlled by the use of solid-liquid cyclone separators,[37] called hydroclones, which fit into the category of mechanical high-temperature fission-product traps.

Gas-Cooled Reactors

A method of keeping the radioactive contamination of the coolant in a gas-cooled reactor at low levels is to confine nearly all the fission-product activity within the fuel elements by the use of suitable cladding. Substantial fission-product contamination should occur only in the event of cladding failure. This approach is employed in the GCR-2 (reference 38) and EGCR (reference 39) designs.

The trend in the more advanced concepts[40–44] is to use fuel elements capable of operating at temperatures of 1800°F or above. The high-temperature gas-cooled reactors are attractive because of the possible achievement of high heat fluxes, high power densities, and high thermodynamic efficiencies, which, in turn, tend to produce both low capital costs and low inventory charges. Metallic cladding capable of withstanding such temperatures does not exist. The refractory materials available at present allow a fraction of the fission products to escape and to contaminate the system. The presence of fission-product activity in the coolant leads to increased shielding requirements for the primary gas system, increased hazards associated with the large amounts of activity circulating in the system, and difficult maintenance problems. Accordingly, the future of the high-temperature gas-cooled reactors appears to be closely associated with the development of methods of controlling and reducing the fission-product activity in the primary gas system. This may be done by the use of relatively impervious fuel elements, by removing the fission products directly from the fuel elements before they can enter the primary stream, or by coolant "cleanup." The extent and mechanism of escape of fission products from some high-temperature fuel elements were discussed in the March 1960 issue of *Nuclear Safety*.[45]

One possibility for the control of fission-product activity is the development of an impervious ceramic coating that would not be subject to deleterious radiation damage. Intensive programs of preparation and testing of advanced fuel elements are currently being pursued at various research installations, and some results are available.[46–49] Detailed discussion of such programs is beyond the scope of this review. It is reasonable to conclude, however, that impervious, properly designed fuel elements constructed of suitable materials constitute the first line of approach to the reduction of radioactive contamination of primary systems of gas-cooled reactors and, in a real sense, are high-temperature fission-product traps.

An alternate approach is to remove the fission products from the fuel elements before they can enter the coolant stream. The concept of fission-product removal from the fuel elements is utilized in an advanced reactor design of the United Kingdom,[44] as well as in a design of the High-Temperature Gas-Cooled Reactor (HTGCR) proposed by General Atomic.[43] One design of a fuel element using this approach consists of a graphite-uranium rod surrounded by a sleeve of low-permeability graphite. A review of fission-product escape from uranium-graphite fuel materials appeared in the March 1960 issue of *Nuclear Safety*.[45]

Several methods of removing fission-product activity from gas streams of reactor systems have been discussed in recent reports.[45–56] The various methods are enumerated here, and some that may be used at high temperatures will be described. In general, different techniques are employed for different chemical species, and the following groups must be considered: noble gases (xenon and krypton), halogens (bromine and iodine), alkali metals (rubidium and cesium), alkaline earths (strontium and barium), and others (yttrium, zirconium, niobium, and selenium).

Methods proposed for the separation of xenon and krypton from helium include the use of in-line charcoal adsorbers,[53–55] molecular sieves,[56]

*The HRT is the aqueous homogeneous reactor now being tested at ORNL.

porous glass,[56] and the formation of clathrates.[57] The schemes mentioned have thus far been used only at low temperatures.

An adequate review of the various methods for halogen removal appeared in the March 1960 issue of *Nuclear Safety*.[52] Some of the more promising methods are reaction with silver-plated metallic mesh or with other heated metals, adsorption by activated charcoal, adsorption by calcium oxide or by soda lime, retention by heated solid silver nitrate, and removal by caustic scrubbing. All these systems, with the exception of caustic scrubbing, may be classified as high-temperature fission-product traps. For the HTGC reactor experiment in England, the use of calcium oxide as a high-temperature adsorber was suggested early.[58] However, soda lime has been found to be more suitable,[51,59,60] and its use is advocated in preference to calcium oxide.

Thermodynamic and fission-yield considerations[61] have indicated the possibility that cesium and rubidium activities might exist in the reactor system as elemental gases and, under certain conditions, as liquid metals. If these activities are not released from the fuel element, they may contribute significantly to pressure buildup within the element which could lead to cladding failure. In the event of escape to the coolant, cesium and rubidium would presumably behave as permanent gases at partial pressures no higher than the vapor pressures of the liquid metals at the lowest temperature of the system.

It has been proposed[61,62] that the addition of some trapping substances (getters) to the system could decrease materially the partial pressures of alkali metals in the gas phase. A list of trapping substances with safe physical and nuclear properties has been reported.[62] The list includes metallic tin and lead, and the addition of bismuth has been suggested.[63] The reduction of partial pressure is achieved by alloy formation or by chemical reaction between the alkali metal and the getter. The extent of pressure reduction will depend either on the composition of the alloy or on the stability of the compound formed. It may be possible to estimate equilibrium partial-pressure values from phase studies and/or thermodynamic tabulations, if available. The actual reduced partial pressures should be measured experimentally under dynamic conditions. Although other methods that may be applicable to the removal of alkali metals include adsorption by activated charcoal and acid scrubbing, no reference to such application has been found.

Alkaline earths and rare earths are also subject to the use of getters, as previously discussed.[62] In addition, the tendency toward carbide formation inherent in the alkaline earths, rare earths, yttrium, and zirconium suggests that these species might be stabilized as nonvolatile compounds. Conversion to carbides could reduce the volatility of these materials by a factor of 10 or more.[63] Experimental determinations of release rates of barium, strontium, cerium, yttrium, and zirconium (initially charged as carbonates, fluorides, or oxides) from hollow graphite cylinders show that, in the temperature range 1200 to 1800°C, yttrium and zirconium have the slowest release rates. Tentative calculations indicate the carbides of yttrium and zirconium to be the most stable.

The removal of solid particles from gases has also been reviewed,[52] and additional discussions pertaining to gas-cooled reactor systems are available.[50,64] The proposed methods involve filtration, the use of cyclone separators, scrubbing, "deliberate suspension of a high concentration of scavenger solids followed by a classical solid-gas separation technique,"[50] and electrostatic precipitation. It appears that the solution of the problem of solid removal must wait until the nature and importance of the problem are better understood from experimental work. (G. M. Watson)

Beryllium Health Hazards

Beryllium is being used in increasing quantities in the nuclear energy program because of its excellent nuclear properties, despite the high cost and the toxicity of the material. It may be used as a moderator or reflector in a nuclear reactor, where a material with low neutron absorption is needed, and it has been proposed for use as a canning material for fuel elements in advanced gas-cooled reactors and in the Project Mercury manned space capsule to take the brunt of the heat during reentry to the earth's atmosphere. This desirable material, even though potentially hazardous to the health of individuals, may be handled safely through proper controls.

Beryllium achieved a bad reputation as a health hazard because of exposures that occurred before it was a recognized hazard. As pointed out in a historical survey of beryllium

disease,[65] the first reports of injury attributable to beryllium came out in the late 1930's and early 1940's. The sources of these reports were plants where beryllium was being extracted from ore. A number of employees who handled beryllium compounds developed a skin rash, and others developed a kind of chemical pneumonia that, in some cases, resulted in death. Three cases of acute "chemical pneumonia" in men extracting beryllium oxide from the ore were described by Van Ordstrand and coworkers in 1943.[65] The disease was characterized by coughing, shortness of breath, and a slight fever. In the same year, Shilin et al. reported disease of the respiratory tract in persons employed in the Pennsylvania beryllium industry.[65] They, however, attributed the illness to the fluoride radical. Two years later, Van Ordstrand reported 38 additional cases of "chemical pneumonia," with five deaths, and 90 cases of mild upper respiratory illness which were attributable to beryllium.

Soluble beryllium salts were thought to be the cause of the cases that occurred in the ore-processing industry. It was postulated that these salts might break down in the body and form acids that would attack the lining of the respiratory passage and lungs. The same disease occurred, however, in people who worked only with insoluble beryllium oxide and metal, and Grier reported[65] seven such cases in laboratory workers. Gradually, through reports from all sections of industry working with beryllium and its compounds, evidence has incriminated beryllium as the causative factor of the illness.

Since 1952, Hardy and her colleagues, as reported by Lieben and Metzner,[66] have collected information on more than 600 cases of all forms of beryllium disease and have established a central beryllium registry at the Massachusetts General Hospital in Boston. In 1959, Chadwick[67] listed over 200 articles related to beryllium health hazards. Therefore there is much information now available, even though most of it has been compiled during the past 13 years.

Medical Aspects of Beryllium Poisoning

The manifestations of beryllium poisoning have been divided into three classes: acute berylliosis, chronic berylliosis, and dermatitis. The most serious of these conditions are entirely due to inhalation of beryllium or its compounds. Acute berylliosis is an acute irritation of the respiratory tract, with pneumonitis, and is usually caused by short-term inhalation of heavy concentrations of beryllium compounds (as low as 50 μg). This type of disease occurs up to one week after exposure, runs its course in a few weeks, and, if not fatal, is usually followed by complete recovery. There are, however, recorded cases in which acute berylliosis later developed into the chronic disease.

Chronic berylliosis has symptoms that are less clearly defined than those of the acute disease. It usually involves loss of weight and appetite, coughing, and shortness of breath. Lung X rays of individuals with the chronic disease indicate, to a trained physician, specific features attributable to beryllium. The symptoms may not appear for as many as 10 to 15 years after the last exposure. Hardy[68] states:

> The most striking feature of beryllium poisoning is a delay between the time of the exposure to beryllium and the onset of symptoms of the disease. In some cases this may extend up to 10 years or longer. Confusion in diagnosis between miliary tuberculosis and chronic beryllium disease may be made by the physician, unless he is aware of the possible effect of beryllium. The patient has great weight loss, cough, shortness of breath, and a chest X-ray much like that seen in forms of tuberculosis. Chronic beryllium poisoning may also involve lymph nodes, liver, spleen, lung, skin, and probably bone.

There is no known cure for chronic beryllium poisoning. The mortality rate of the chronic disease is at present 20 to 25 per cent, and the Massachusetts General Hospital data show complete invalidism in 50 to 60 per cent of the cases.[69]

Contact dermatitis[70] may occur in people working with beryllium fluoride within a few days of their first exposure to either dust or fume; it is often accompanied by conjunctivitis, and it heals upon cessation of exposure. Other soluble beryllium compounds appear to have the same effects. Penetration of cuts or abrasions by solid crystals of such compounds may lead to painful ulcers, which persist until the beryllium is removed.

Preliminary medical examinations are necessary before permitting personnel to work with beryllium, and periodic examinations, including chest X rays, should be continued. As for urine analysis, Hardy[69] says, "Beryllium in urine is an index of exposure only and not related to present disease or possible future illness."

Hazard Control Criteria

Recommendations for the control of beryllium hazards have been published by the USAEC,[71] and similar controls based on these recommendations have been suggested in Great Britain,[72] as mentioned in the June 1960 issue of *Nuclear Safety*.[73] At the risk of being repetitious, a more complete statement of the U. S. recommendations is presented below:

1. The in-plant atmospheric concentration of beryllium at beryllium operations should not exceed 2 $\mu g/m^3$ as an average concentration throughout an 8-hr day.

2. Even though the daily average might be within the limits of recommendation 1, no personnel should be exposed to a concentration greater than 25 $\mu g/m^3$ for any period of time, however short.

3. In the neighborhood of an AEC plant handling beryllium compounds, the average monthly concentration at the breathing-zone level should not exceed 0.01 $\mu g/m^3$.

4. There should be an adequate medical program, supervised by a physician who is familiar with beryllium poisoning, to cover all workers exposed to beryllium and its compounds.

5. If there is any evidence that an individual has chronic beryllium poisoning, such an individual should be excluded from any further exposure to beryllium compounds.

6. Beryllium operations should be carried on in as few places as possible.

7. Beryllium and beryllium compounds should not be used by uninstructed or unsupervised groups in any operation that will produce atmospheric contamination (dusts, fumes, mists, etc.).

Plant Control of Beryllium Hazards

Adequate provisions may be made in any facility to achieve safe working conditions based on the information cited above. The required provisions consist of a combination of measures, including monitoring, ventilation, contamination control, and personnel education.

Detection of low-level beryllium concentrations is required as quickly as possible in order to evaluate conditions. Instantaneous sampling, analysis, and recording would be desirable but to date have not been practical for routine operations because of the size and maintenance requirements of instantaneous beryllium monitors. Samples are usually obtained by pulling measured amounts of air through filter paper that will retain particles down to 0.2 μ in diameter. The particles on the filter paper are then analyzed by the method most practical for the particular plant. Spectrographic analysis of the material impacted on the paper seems to be most popular. Such analyses can be made rapidly, and the sensitivity of the method is good. Smears or swipe samples are generally taken for control of housekeeping, contamination level of outgoing equipment, etc. The limits used for such contamination vary from one installation to another, with the goal being to keep surface contamination from accumulating enough to increase air-sample results.

Ventilation control is required to minimize beryllium contamination. In most facilities the flow of ventilating air is from the outside to the room or rooms involved, and the air is exhausted through machine hoods, laboratory hoods, etc. Recirculation of air within a beryllium-processing area is not recommended. All exhaust air should pass through absolute filters that will remove submicron-size particles to prevent the spread of beryllium contamination into building air intakes and neighboring areas. The number of air changes in a room may vary from 5 to 40 per hour, depending on the operations.

All processing should be performed, if possible, in enclosed systems with the maximum of automation. Since not all operations can be performed in this manner, most authorities agree that a minimum air velocity of 150 ft/min across the processing area is needed to maintain good control. The extent of the enclosure will depend on the operation and may vary from an open hood for wet handling and less hazardous work to a glove box for handling fine, dry powders. High-velocity (600 to 1000 ft/min) exhaust air has been used successfully during dry machining by mounting the opening of a flexible air tube at the point of cutting. When a liquid coolant is used, the fluid spray must be controlled, and good housekeeping is a necessity.

Beryllium or its compounds in powder form should not be handled in open hoods that communicate with an unfiltered ventilation system. Dry sweeping should not be permitted; however, an oiled sweeping compound may be used. A vacuum cleaner with an outlet filter that retains 0.2-μ particles is approved for beryllium areas. All materials leaving a beryllium area should be thoroughly decontaminated and checked by a swipe test. General washing and cleaning of the

area should be done on a routine basis and at other times when a swipe test indicates that such cleaning is needed. Large amounts of liquid or solid residues or unrecoverable contaminated equipment should be sealed and transported under controlled conditions to a designated area for burial.

It is the consensus that all personnel working with beryllium should be provided a complete change of clothing for each shift and required to shower at the end of each shift. Clothing should be wetted down after removal and laundered under controlled conditions.

Conclusions

It has been conclusively demonstrated that beryllium is hazardous to the health of personnel when inhaled. The British appear to place more emphasis on processing beryllium in enclosures, such as glove boxes, than American industry does, outside the AEC, but both agree that the limits that have been established are safe. Although the consequences of either the acute or chronic disease may be lethal, and over 50 per cent of the chronic cases result in complete invalidism, present control criteria, if faithfully followed, are sufficient to eliminate virtually all instances of exposure. From the time of the adoption of the standards for control of beryllium in 1949 to June 1958, only 10 cases of beryllium poisoning, unrelated to exposure before 1949, were reported in the United States, and none were reported in Great Britian.[69] It may be concluded that beryllium can be handled safely through proper precautions involving particularly the control of airborne contamination, since the greatest hazard is inhalation of the material.

(J. F. Morehead and F. Williams)

Fuel-Processing Hazards

The design and construction of facilities for processing first-generation power-reactor fuels require the solution to a number of new and especially difficult radiation and criticality control problems. These problems are occasioned by the properties of the fuel elements, of which there are many types, and by the desire for development of processing equipment that can economically process fuel at rates which are many multiples of the minimum critical mass per day. The power-reactor fuel elements now being designed and fabricated are characterized by high radioactivity and high fissile-material content, and, although manufactured by a variety of techniques, they consist of stable, corrosion-resistant cladding and core materials.

Processing problems have been studied for the past several years at the various AEC processing sites, since the AEC plans to process power-reactor fuel until such service is available from private industry. The proceedings of a recent symposium[74] describe the results of some of these studies and the alternate methods of radiation and criticality control which have been evolved. Earlier information on this subject was given in the December 1959 issue of *Nuclear Safety*.[75] Some of the most significant hazards that have been evaluated are (1) containment hazards occurring in the receipt, mechanical treatment, and storage of fuels; (2) criticality hazards occurring in dissolution and solvent-extraction feed preparation; and (3) explosion hazards caused by potentially explosive off-gases.

Fuel Handling and Storage

At ORNL, the Idaho Chemical Processing Plant (ICPP), and the Savannah River Plant (SRP), water-filled canals are planned for use in unloading, mechanical alteration (removal of inert end sections, etc.), and storage of power-reactor fuels.[74] Canals have been used successfully for this purpose with production and test fuels. Water has the advantage that it constitutes an excellent shield for both gamma radiation and neutrons and allows good viewing for the handling operations. Its disadvantages are that it does not constitute good containment for flying fragments or gases which might be formed or released in chemical or nuclear explosions. At the processing sites that plan to use canals, the canals are to be enclosed within a building that will serve to minimize escape of radioactivity to the environment in the event of an accident. Contamination of the canal water will be minimized by "canning" ruptured fuel elements and by periodic or continuous repurification of the water.

The processing plant at Hanford Atomic Products Operation (HAPO),[74] on the other hand, will handle and unload all irradiated power-reactor-fuel shipping casks in air. Casks will be transferred by direct operation from cross-country railway cars or trucks to a plant railway car in

a specially constructed transfer building. The plant railway car will then be drawn into the potentially contaminated, shielded canyon, where the fuel elements will be remotely unloaded from the cask and placed in water-filled "bathtubs." After fuel elements are removed from a shipping cask, the cask will be withdrawn into the transfer building, where it will be decontaminated by direct contact prior to its return to the shipper.

The HAPO fuel-element-storage bathtubs will be located within separately shielded storage cells within the canyon. The normally stagnant water within the bathtubs, which will be heated by the decay heat in the fuel elements, will be maintained below 150°F by cooling-water flow through coils within the walls of the bathtubs. This cooling water will be discharged directly to the environment. The normally stagnant water within the tubs will be carefully monitored to detect the presence of any radioactive material that may have leached from a fuel element, and provisions will be made to reclaim any fissionable material in the water.

Another type of cell to be provided within the canyon will be equipped for remotely removing nonvaluable components from the fuel elements and reducing the fuel elements to a size suitable for charging to a dissolver that is geometrically safe nuclearwise. This mechanically equipped cell will also serve a nuclear safety function in that inert residues from dissolving operations will be remotely examined within the cell for fissile-material content prior to disposal. This is a straightforward, but unique, method of avoiding the nuclear safety and accountability problems caused by undissolvable residues. All cutting of potentially pyrophoric materials, such as zirconium, will be done under water, and the water will be recirculated through a filter to recover the fines.

The air flow in the ventilation system for the canyon, storage cells, and mechanically equipped cell will be from clean areas into progressively more contaminated areas. Exhaust fans will pull the ventilation air through inlet filters, into the canyon, through cracks in the cover blocks into the cells, and from the cells through sand filters, and the air will discharge through a 200-ft-high concrete stack. The ventilation air for a mechanically equipped cell will also pass through a water scrubber ahead of the sand filter. It is stated that the sand filter is better than 99 per cent efficient in removing submicron particulate matter encountered in normal plant operations.

The proposed HAPO method of remotely receiving, mechanically treating, and storing fuel elements within shielded concrete cells provides a higher degree of containment than water canals. This type of containment is, however, considerably more expensive than the apparently suitable canals. Selection of the best of the two procedures for processing plants to be built in the future must depend on the demonstration of the safety of water canals in processing high-level power-reactor fuels.

Dissolution and Solvent Extraction

The selection of methods for criticality control during dissolution of power-reactor fuels and preparation of solvent-extraction feed at the various processing sites depends on the type of fuels assigned to the site and the facilities presently available. HAPO and ORNL have chosen to use, primarily, two-step dissolution processes on the fuels they have been assigned. The processes consist of dissolution of the cladding with a suitable aqueous reagent, removal of the cladding solution from the dissolver to the waste system, and addition of another reagent to dissolve the core material, followed by removal of this solution from the dissolver to a solvent-extraction feed tank. These processing steps are characterized by a criticality hazard, particularly when used with fuel elements containing ceramic core materials, because solid fuel fines may be carried by the cladding-material solution as well as by the core solution. Adequate solid-liquid separation and detection methods must therefore be devised.

The proposed Hanford method[74] of solving the nuclear safety problems in these so-called "head-end" processes for fuel with up to 5 per cent enrichment will be to use a geometrically safe batch dissolver constructed of combinations of 7.8-in.-diameter cylinders and 2.9-in.-thick slabs. All solutions that leave the dissolver will be routed through geometrically safe piping to a geometrically safe centrifuge, possibly a continuous liquid and solid discharge type, for removal of possible fissile-material-containing solids. The supernatant liquid from the centrifuge will be collected in geometrically safe tanks, analyzed, diluted to always safe concentrations, and passed to nongeometrically safe storage tanks prior to solvent extraction.

The proposed ORNL method[74] of head-end criticality control utilizes a semicontinuous dissolver that is geometrically safe with respect to the batch in the dissolver followed by geometrically safe gravity settlers for removal of fissile solids and by appropriate valving for discharge of cladding solution or core solution to large catch tanks. The large catch tanks have geometrically safe bottom sections equipped with means for detection of accumulated fissile solids. The solution of fissile material in the catch tank is to be rendered safe by the use of a sufficient quantity of soluble neutron poison, whose presence is guaranteed by procedural, analytical, and instrumentational safeguards. A neutron absorption monitor is to be used for in-line measurement of the poison content of solutions.

Three instruments[76] are being considered for use in detecting the presence of fissile and fission-product-containing solids in the geometrically safe bottom sections of catch tanks. They are gamma-ray monitors, neutron multiplication monitors, and specific-gravity determination devices. The gamma-ray monitor, equipped with an ion chamber, would indicate the presence of irradiated fissile material by detecting the increased gamma-ray field which must accompany an accumulation of irradiated fuel. Precautionary measures (redundancy, shielding, etc.) would be expected to assure a useful life for the monitor of greater than one year. The neutron multiplication instrument can detect buildup of fissionable material even when installed in high-background-activity areas. Shielding of the BF_3 counter to a level of 10 r/hr is required, but this can be done without interfering with the accuracy of the instrument. Solids, if collected into a small volume, can be detected by variations in specific gravity of the solution during periods of agitation.

The apparent chemical feasibility of using boron, cadmium, or rare-earth salts as nuclear poisons has been demonstrated and is reported in an ORNL report.[77] These poisons are soluble and stable under process conditions in sufficient quantity to poison concentrations of fissile isotopes of 100 g/liter or more. None of these poisons were volatilized to a significant extent in any of the process treatments, one of which included evaporation at a temperature of 140°C. It was also demonstrated that each of the poisons could be adequately separated from fissile and fertile product material by solvent extraction.

A geometrically safe centrifuge is also being considered for use at ORNL.[76] The preliminary design comprises an annular modification of a conventional, continuous-liquid-discharge, batch-solid-discharge centrifuge.

At SRP[74] it is planned to dissolve four Power Reactor Development Company (PRDC) fuel elements which contain approximately 75 kg of 25.6 per cent enriched uranium in a geometrically safe basket in a large-diameter dissolver with essentially a complete batch-dissolution process. The fuel elements in the basket will be dissolved in sufficient acid to provide an always safe concentration of 9 g of U^{235} per liter in the large-diameter vessel. The fuel insert and dissolver steam coils are mounted high in the dissolver so that, if the solution inadvertently boiled below the level of these heat sources, the solution volume would be such that the batch of fissile material would remain below the critical concentration. Instrumentation is used to monitor the fissile concentration in the solvent-extraction plant. Instruments now in use for this purpose include conductivity cells, colorimeters, and neutron multiplication monitors.

Disposition of Off-Gases

HAPO and ORNL have described alternate methods for safely disposing of the H_2 and NH_3 gases which will be generated in decladding processes.[74] At HAPO, sweep air will be provided in the dissolver in sufficient quantities to lower the quantities of H_2 and NH_3 to below explosive limits all through a standard dissolver off-gas train. This essentially serves to multiply the volumetric flow rate of the H_2 and thus the cross-sectional area of piping, filters, and scrubbers by a factor of 25 to 50. Therefore a small off-gas stream that is undiluted with large quantities of sweep air is desired to minimize the size and thus the cost of the off-gas removal equipment. This will be accomplished by maintaining the O_2 content of the H_2- or NH_3-containing gas streams below the minimum required for combustion. The dissolver and off-gas system will be maintained relatively leaktight and at a negative pressure with respect to the atmosphere to prevent leakage of potentially explosive gas to the cells. In the event that accidental inleakage of O_2 should occur at a high

rate, N_2 will be added to the system automatically by equipment actuated by an O_2 analyzer.

The British described[74] safeguards that will be incorporated in new chemical separations cells which will be used to reprocess irradiated power fuel. The cell ventilation system will be arranged so that air is drawn from the operating area into the cells, from the cells through filters, and then exhausted from a stack by fans. The ventilation flow will be such that an air velocity of 3 ft/sec will be maintained through all penetrations in the biological shield. Fire control in the cells will be achieved through the use of CO_2 as a smothering gas. The CO_2 is stored in liquid form at high pressure and is coupled to a detection system that automatically releases the gas to an affected cell in the event of fire.

Tanks for Liquid Storage

The reviewer has studied the applicability of large-diameter vessels packed with boron-containing glass raschig rings for storage of high-concentration solutions of fissile material. It is calculated that 3-ft-diameter 250-gal vessels packed to 11 vol.% with 1½-in.-diameter glass raschig rings containing 3.9 wt.% boron would be critically safe for storage of U^{235}, U^{233}, and Pu^{239} solutions of 100 g/liter. Vessels of the same size packed to 25 vol.% glass with rings containing 6 wt.% boron may possibly be safe for solutions of these isotopes regardless of the solution concentration. Use of this means for storage of fissile solutions could result in appreciable cost savings in comparison with the cost of geometrically safe storage vessels. Information from Alcorn and Mottley[78] (as yet unpublished) summarizes the results of mixing, corrosion, and stability experiments that were performed with a 250-gal tank packed to 22.6 vol.% glass with 1½-in.-diameter glass raschig rings containing 6 wt.% boron. It was determined that 150 gal of solution within the tank could be homogeneously mixed in approximately 30 min by an air sparge ring operating at an air flow rate of 0.55 scfm per square foot of tank cross-sectional area. Limited data indicate that boron may be leached from the glass rings by $2N$ HNO_3 at room temperature at rates of 0.1 to 0.2 wt.% boron per year. No cracking or settling of the rings within the tank was observed during the one-week period in which the tests were performed. (J. P. Nichols)

Decontamination Hazards

In an earlier review of decontamination hazards,[79] it was pointed out that the hazards during decontamination of radioactive equipment may be greater than during normal operations and, hence, should receive special consideration. As before, the scope of this review includes only criticality and radiation hazards; no attempt is made to cover "ordinary" hazards such as chemical effects, heat, and pressure. Although a decontamination task may be sufficiently urgent to require the acceptance of hazards, the hazards and risks involved must be recognized and decisions made on the basis of all circumstances involved, including the necessity for achieving the maximum degree of safety consistent with the situation.

Radiation Hazards

Actual experience in an emergency decontamination situation is of particular interest in a discussion of decontamination hazards. The decontamination task that followed the NRU reactor incident[80] in May 1958 has been described by Hughes and Greenwood.[81] The contamination resulted from efforts to remove a ruptured fuel rod from the reactor. During removal operations, cooling water was lost and the fuel ignited. In transporting the fuel rod, a 3-ft section dropped into the maintenance pit "began to burn and set fire to paper and other materials there." The radiation level around and over the maintenance pit was greater than 1000 r/hr. The fire was smothered by wet sand carried in buckets within 15 min of ignition, and the highest exposure in this operation was 5.3 r. Next, the burned rod segment was removed by "a team of men, working 1 min at a time and using long-handled tools (rakes, hoes, and shovels with 24-ft handles)" into a wooden tray filled with sand. "Lifting the tray into the maintenance pit and transferring it to the (shielded semi-trailer for transportation to the disposal area) were difficult. Radiation fields in the cab of the overhead crane ... were 5 to 10 r/hr, requiring a change of operators every 2 min."

"All the operations described so far were carried out by about 35 men. Despite the fact that they worked near fields up to and over 1000 r/hr, the average radiation dose received was only 1.4 r, and the maximum individual exposure was only 6.4 r. This record indicates that ex-

treme care was taken in timing each exposure and that the men concerned took advantage of all available shielding." Additional men, "whose normal work did not involve exposure to radiation, under the direction of an experienced employee who remained shielded," were assigned for further cleanup work. Forty men, working in pairs, removed the sand from the pit. "... in spite of the 1½ min working time prescribed for each man, 14 of them exceeded the 5 r limit. Of the latter, 11 received 5–6 r (probably by lifting isolated bits of uranium in the sand), two received about 10 r (by slightly overstaying their allotted 1½ min), and one received 19 r (by making an extra trip to the pit edge ...). These exposures were the highest received during the entire cleanup." Presumably the 1½-min working time was calculated as the time during which the dosage would not exceed 5 r. Since some exposures were considerably in excess of 5 r, it would appear that in this phase the supervisors were not so careful and/or the additional men had not been so well indoctrinated as in the preceding phase.

Many details of the decontamination procedure are given,[81] such as the fact that 25,000 rubber overshoes were decontaminated in July 1958. Tables are presented showing contamination levels and the number of exposures at various dose ranges. Some information is also given on personnel protection. "Each man employed in the cleanup was provided with protective clothing (disposable plastic suits), a full-face respirator, a film badge and a pocket dosimeter. Dressing and undressing were carefully supervised by experienced personnel .... Rubber gloves, respirator and rubber overshoes were sealed to the suit with tape. In the early stages of the cleanup, it was not unusual to find as much as 200 mr/hr surface contamination on used suits. By taking special care to undress the wearer by turning the garment inside out, very few cases of skin contamination were encountered."

Considering the nature of the accident, it would seem that the internal radiation hazard approached the external hazard in seriousness. "During the first night of the emergency, protection against internal contamination was not as effective as it was in the later stages. Men present while the uranium was burning were dressed in white coveralls and were wearing modified 'Comfo' dust masks, which cover the mouth and nose .... Some cases of internal contamination were traced to improper use of respirators—for example, raising the face piece briefly to breathe more easily or to speak to another worker .... Other cases resulted from the relative ineffectiveness of the dust masks against iodine. Nevertheless, the overall control of internal contamination was highly effective." The authors[81] reach this conclusion on the basis of 2000 urine samples, of which less than 10% were positive for mixed fission products or iodine. "Subsequent calculations showed that no one had received an overexposure to radiation as a result of internal contamination." It is not clear whether these calculations were based on internal exposure alone or on internal plus the external exposure received by the individual, but, in either case, control of internal contamination does not appear to have been as effective as desirable.

During the NRU decontamination operation, a technique was used which undoubtedly significantly reduced the personnel exposure and hazard.[81] "Through the use of a remote-controlled television system, it was possible to view (from an adjacent building) nearly all areas in the reactor hall and, through the use of the reactor public-address system, to give instructions to personnel actually engaged in the cleanup. The television system was particularly useful for showing workers exactly what their duties would be when they entered the reactor hall."

Of particular significance is the statement:[81] "Much more contamination was spread on roads used for transportation of debris from the uranium fire than escaped directly from the building. The roads were cleaned quickly by vacuuming, washing with fire hoses, and, when necessary, removing part of the road surface." This experience should serve as a warning that careful planning is necessary to ensure that decontamination results in removing a hazard rather than merely changing its location.

Another major decontamination program was necessitated by the Nov. 20, 1959, activity release at ORNL. This activity release, previously described in the March 1960 issue of *Nuclear Safety*,[82] resulted from a chemical explosion in the Thorex Pilot Plant and involved approximately 15 g of plutonium, of which about 0.6 g escaped from the building. The immediate hazard from contamination outside the building was eliminated by such measures as spray painting to "fix" the plutonium and removal of a contaminated section of roadway. From this point on, as contrasted with the NRU cleanup,

decontamination of the pilot-plant building (Building 3019) proceeded at a very deliberate pace and, as of August 1960, was still in progress in some areas of the building.

In cells 6 and 7 where the explosion occurred, alpha activity smears counted as high as 10^8 dis/min per 100 cm^2. Outside these two cells, the highest smear counts were 50,000 dis/min per 100 cm^2, and direct alpha activity probe measurements were as high as 800,000 dis/min per 100 cm^2. For clearance of any area for personnel occupancy, the following alpha-activity contamination specifications were set in terms of disintegrations per minute per 100 cm^2 of surface area:

	Uncontaminated areas	Contaminated areas
Direct probe reading:		
Maximum	300	3000
Average	≤30	≤300
Transferable (smear):		
Maximum	30	300
Average	≤3	≤30

In a contaminated area, after these specifications were met, the remaining contamination was fixed with a bond, such as a color-coded paint (under coat orange or red to signal the need for repainting if the top coat flakes or wears thin) or concrete. Protective clothing is always required in a contaminated zone, even though the contaminant is sealed under paint or concrete. It is the opinion of the reviewer that the specifications outlined above (particularly those for contaminated zones) should be reexamined to determine whether they are more restrictive than necessary.

King and McCarley have described some of the precautionary measures used in the cleanup:[83]

> Before actual decontamination effort was begun, it was necessary to set up strict rules for entering and leaving contaminated areas to prevent the spread of contamination into clean areas. Also, maximum protective clothing requirements for contaminated zones were designated as two (2) sets of coveralls, two (2) pairs of shoe covers, two (2) pairs of rubber gloves, assault masks, hood, and wrists and ankles taped. This was required in all contaminated zones except in Cells 6 and 7 where plastic air suits were required. When leaving contaminated zones, clean-up personnel were directed to well defined check points where successive layers of clothing were removed and personnel were monitored. Personnel then showered and received a final check by Health Physics surveyors before being released. Frequent smearing of the change house area indicated that little or no contamination was spread into clean zones.
>
> Before decontamination work was begun on each area, easily removable equipment was carefully wrapped in plastic and moved to an outside decontamination pad. Equipment and supplies of low value were removed to the "burial ground" for disposal.

The effectiveness of the control measures employed is illustrated by the exposure record since the explosion. Data that will be published as Report ORNL-2989 show that no one received an overexposure, either internal or external. During the decontamination program (through June 1960), the highest three-month external exposure for any individual was 705 mr. Three other people received dosages in excess of 600 mr in a quarter, and four received dosages between 500 and 600 mr; all others were under 500 mr per quarter. The six-month external exposures (including the quarterly exposures mentioned above) were: one man, 1070 mr; three men, 900 to 1000 mr each; three men, 800 to 900 mr; and nine men, 700 to 800 mr; the remaining men have received less than 700 mr in six months. There were no internal overexposures. The two men who received the highest internal exposures were exposed to plutonium plus zirconium-niobium activity. One of these men was an operator who was exposed the night of the explosion, and the other was a lead burner who was exposed during decontamination. In both cases, the maximum permissible concentration in the lungs was the controlling factor, although the total systemic hazard was almost as great. Since there were probably localized exposures within the pulmonary system, an assumption of uniform distribution within the lungs was not necessarily valid, but whole-body counting indicated reasonable uniformity. Assuming uniformity, both lung dosages would result in long-term storage in the bone of about 2 per cent of the maximum permissible systemic body burden of plutonium.

One sidelight of these decontamination operations may be indicative of future difficulties for other installations. The building was originally constructed as a pilot plant for bismuth phosphate processing and, since 1944, has been used as a pilot plant for several irradiated-fuel processing systems, such as Redox and Purex. In some areas, successive attempts at decontami-

nation, including concrete chipping, did not appreciably decrease the level of activity. Analyses of the contaminants showed that the material deeply embedded in the concrete or adhering tenaciously to stainless steel did not come from the explosion but, rather, had remained from earlier programs. The areas of the building which have been decontaminated and cleared for operational use are probably freer from contamination now than they have been at any time in the past 15 years.

There are several other reports of interest. Especially pertinent is a discussion by Goren and Thornton[84] of radiation hazards in ship repair, since the repair procedures they discuss, i.e., sandblasting, welding, chipping, disk sanding, and wire brushing are standard decontamination methods. A series of tests, "modest in scope and more qualitative than quantitative in nature, ... were performed on a vessel which had participated in recent nuclear tests and which still had moderate-level radioactive contamination." The measurements showed primarily beta activity, and it is stated that the measurements "may require an additional correction factor." The readings are comparable, however, and the ratios are valid. The air in the test area was sampled by filtering through CWS No. 5 paper "with a particle capture efficiency of 80 to 90%."

Data collected for each of the repair procedures are presented in both tabular and graphic form in the report by Goren and Thornton.[84] Corrected beta-activity surface intensities ranged up to 29.5 mrads/hr. The work-area air was sampled at various distances from the work and under varying conditions of air movement. The air activities are reported as the fraction of, or the number of times greater than, the maximum allowable concentration (unprotected). Although the data on sandblasting are not directly comparable to the other measurements, the data make sandblasting appear to be the most hazardous method tested, with dry blasting being worse than wet blasting. The other procedures, in decreasing order of severity, are disk sanding, pneumatic wire brushing, welding, and pneumatic chipping. The authors conclude that "by using local exhaust ventilation and high-efficiency filter type respirators, any respiratory hazard to the operator can be eliminated when dusts are produced in working on surfaces having low-level radioactive contamination."

The term "low-level" is critical, since the data show not only distances and intensities at which no protection is needed, but also conditions under which a filter is inadequate and a clean air-flow mask is required. For example, in disk sanding on a surface whose corrected beta-activity intensity is 20 mrep/hr, the airborne contamination at zero distance is 65 times the maximum allowable concentration; at 8 ft it is 10 times the maximum allowable concentration; and only beyond 12 ft is it less than the maximum allowable concentration.

Two pertinent reports from the Naval Radiological Defense Laboratory were also reviewed. The first of these[85] deals with "the application of radiological safety procedures in the event of a nuclear weapons accident." The report outlines the procedures and gives frequent reference to other publications, primarily U. S. Navy publications, for details. It also outlines personnel protective measures, such as provisions for clothing, personnel monitoring, showers, respiratory protection, and dosimetry devices. A warning in the report further illustrates the point made above that decontamination may result in merely transferring the location of the hazard. It is stated that "at the time of the accident and subsequent actions, contamination is liable to reach the sewage system. Settling points in such a system and treatment facilities should be monitored for contamination by liquid and sludge sampling and decontaminated as necessary by sludge removal and flushing."

It is pointed out that "certain recovery operations may increase the airborne contamination by resuspension of surface contamination." The authors give a "resuspension factor" (4×10^{-8} $\mu c/cc$ per $\mu c/cm^2$) which can be used to estimate the airborne contamination from a known surface contamination level of plutonium (presumably as the oxide). Both the range of applicability and the validity of this resuspension factor are highly questionable even for order-of-magnitude estimation. However, if such a factor (or even table of factors covering different conditions) could be validated, it would be of great value for advance estimates of the hazards of decontamination of a surface.

The other Navy document[86] describes tests of dry methods for cleaning paved areas. The equipment utilized in the tests consisted of an ordinary "motorized" street sweeper that was operated dry and was equipped with a conveyor and a hopper for collecting debris, a "vacu-

umized" street sweeper with a cloth-bag filter, and an "air broom" which comprised, essentially, compressed air nozzles for blowing dirt off the street. The tests were primarily an evaluation of effectiveness and of effort required. All the methods tested appeared to spread contamination. The hopper on the motorized sweeper leaked, particularly when nearly full, and probably stirred up small dust clouds. Although it was not stated, cloth-bag filters undoubtedly have a low collection efficiency for small particles and could cause recontamination and an airborne hazard. "The removal of heavy deposits by the air broom produces a large dust cloud." Nevertheless, the authors recommend that the methods be adopted as: "(1) a supplement to wet methods and (2) in areas of critical water supply, as the primary decontamination method for paved areas." It is obvious that these methods are hazardous, but the fact thay they are still recommended points up the need for a balance of the hazards with the circumstances involved. It is difficult to conceive of many situations (short of clearing a route for an emergency evacuation) in which such hazardous methods would be justified, but it is possible to picture certain military situations in which circumstances would necessitate the acceptance of the hazards.

Criticality Hazards

No specific discussion of criticality hazards during decontamination has been found in the literature, but several decontamination procedures have been proposed[87–89] in which the collection of a critical quantity of fissionable material is conceivable, even though highly improbable. For example, a complexing oxidizing decontaminant is proposed for removal of UO_2 (natural uranium) in the Shippingport power plant.[87] Even though the solution was developed for decontamination following a blanket fuel-element failure, the statement is made that the solution "is suitable for in-pile loop decontamination after major UO_2 or high-uranium alloy fuel failures." There is always a possibility that a procedure which is inherently safe in the application for which it is designed may be applied to a system in which there exists the possibility of additional hazards.

Another example is the basic permanganate-citrate procedure proposed for decontamination of the Yankee reactor primary cooling system.[89] Uranium dioxide is insoluble in the decontaminants to be used, but the possibility of the formation of a slurry must be considered. In addition, there have been proposals to adopt this procedure for reactors that use metallic rather than oxide fuel, and, in this case, solubility must be considered.

As mentioned before,[79] even though criticality hazards are very improbable during decontamination, the possible consequences of a criticality incident are so serious that criticality hazards must be specifically considered in any decontamination plans. In addition, any proposal for a decontamination method should include at least a statement of the criticality factors considered in order to avoid extension of the method to an unsafe system. (W. H. Carr)

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Section V

CONSEQUENCES OF ACTIVITY RELEASE

Atmospheric Dispersion Calculations Using the Generalized Gaussian Plume Model

By F. A. GIFFORD, Jr.

A number of formulas for dealing with various practical dispersion problems that arise in reactor hazard analyses are based on the widely used dispersion model formulated by Sutton[1] and are listed[2] in USAEC Report AECU-3066, Meteorology and Atomic Energy. As pointed out in a previous article by the author in the March 1960 issue of *Nuclear Safety*,[3] however, results of recent dispersion experiments have more and more often been presented in terms of the following simple Gaussian interpolation formula:

$$\chi = \frac{Q}{\pi \sigma_y \sigma_z u} \exp\left[-\frac{1}{2}\left(\frac{y^2}{\sigma_y^2} + \frac{h^2}{\sigma_z^2}\right)\right] \qquad (1)$$

where χ = concentration in grams or curies per cubic meter
Q = source strength in grams or curies per second
u = mean wind speed in meters per second
y = crosswind in meters from the plume axis, which is assumed to coincide with the mean wind direction
h = source height in meters
σ_y^2, σ_z^2 = dispersion coefficients in square meters

The right-hand side of Eq. 1 contains a multiplicative factor of 2, which is a conventional device to account for the assumed reflection of the plume by the ground plane. The relation of this formula to Sutton's dispersion model is given in the earlier article by Gifford.[3] The earlier article indicates that, in view of the considerable theoretical interest in the Gaussian formulation, there appears to be a practical need for a corresponding group of special formulas based on Eq. 1.

Volume-Source Formula

The need for modifying Eq. 1 for the case of a volume source arises primarily in connection with the problem of emission of airborne radioactive material through possible leaks in a reactor containment structure. In a reactor hazard analysis the source is generally shown to consist of some fraction of the fission products contained in the reactor core, and the source material is assumed to be distributed uniformly throughout the volume of the reactor building. For many power reactors the enclosure is a large pressuretight dome designed to have, at most, some specified leakage rate under the postulated accident conditions. The source strength Q is defined, but the location of the leak and the effect of the building on the source geometry must be determined.

Reasoning that a reactor building must have a turbulent wake in its lee, Fuquay[4] suggests treating the building effect as an initial dilution factor D_B where

$$D_B = cAu \qquad (2)$$

and A is the cross-sectional area of the building normal to the wind. It is estimated that $\frac{1}{2} \leq c \leq 2$, i.e., any material escaping from the containment building is asumed to be dispersed rapidly into a volume equal to c times the building cross-sectional area times the wind speed. The factor c represents an estimation of the relation of the building cross-sectional area to

the size of observed pressure wakes, and its exact numerical value will have to be determined by suitable experiments.

The building dilution factor, D_B, is combined with the atmospheric dilution factor, $D_A = Q/\chi$, in a manner similar to Fuquay's[5] handling of stack dilution:

$$D_{\text{total}} = D_B + D_A \tag{3}$$

By combining Eqs. 1, 2, and 3, it follows that, in the downwind direction ($y = 0$),

$$\chi = \frac{Q}{(\pi\sigma_y\sigma_z + cA)u} \tag{4}$$

where h is (conservatively) assumed to be equal to zero.

The Fumigation Condition

Hewson[6] introduced the term "fumigation" to describe the mixing downward to the ground of material which has accumulated aloft during a period of atmospheric stability; such an occurrence is common after dawn, when the nocturnal temperature inversion is rapidly dissipated by warming due to solar heating of the ground. Concentrations due to the fumigation effect are estimated[2] by integrating Eq. 1 with respect to z from 0 to ∞ and then assuming that the cloud material is distributed uniformly through a layer of height H. For example, H could be the height of the base of a persistent inversion aloft, such as the West Coast (California) subsidence inversion. Alternatively, H might be the depth of the planetary boundary layer, i.e., the haze layer. Scorer[7] states that a stable layer approximately equal to the elevation of ridge tops often marks the upper boundary of smoke diffusion in valley locations; H could be identified with this level. The equation for the fumigation concentration, χ_F at $y = 0$, is

$$\chi_F = \frac{Q}{(2\pi)^{1/2}\sigma_y uH} \tag{5}$$

Crosswind Integrated Concentration

The crosswind integrated concentration, χ_{cic}, is obtained by integrating Eq. 1 with respect to y from $+\infty$ to $-\infty$, as follows:

$$\chi_{cic} = \frac{(2)^{1/2}Q}{(\pi)^{1/2}\sigma_z u} \exp\left(-\frac{h^2}{2\sigma_z^2}\right) \tag{6}$$

This equation is particularly useful as an interpolation formula in field dispersion trails because it contains only one dispersion parameter, σ_z.

Long-Period Average Concentration

Over a period of time the direction of the mean wind shifts. The wind rose, which gives the joint wind-speed and wind-direction frequency distribution, is therefore a useful indicator of the characteristic features of the climate in a particular locality. To obtain an estimate of the average concentration over a period of time that is very long compared with that over which the mean wind is computed, the crosswind integrated concentration formula given in Eq. 6 is multiplied by the frequency, f, with which the wind blows toward a particular location during the period; the resulting χ_{av} at a point, for f in per cent per radian, is

$$\chi_{av} = \frac{(2)^{1/2}0.01fQ}{(\pi)^{1/2}\sigma_z u} \exp\left(-\frac{h^2}{2\sigma_z^2}\right) \tag{7}$$

This expression forms the basis for the calculations by Meade and Pasquill,[8] who computed annual SO_2 concentrations in the vicinity of the Staythorpe, England, power station (using the corresponding Sutton formula).

Maximum Concentration and Its Distance from the Source

Because σ_y and σ_z are not necessarily the same functions of x, it is not generally possible to obtain simple, explicit formulas for the maximum ground concentration and its distance from the source. However, in the special case $\sigma_y = \sigma_z$, which indicates neutral or slightly unstable conditions, these maximum values can be specified. Differentiating Eq. 1 with respect to x and setting the result equal to zero gives

$$\chi_{\max} = \frac{2Q}{\pi h^2 eu} \tag{8}$$

when $h^2 = 2\sigma_z^2$. In the slightly more general case, which is characterized by $\sigma_y = a\sigma_z$, $d\sigma_y/dx = ad\sigma_z/dx$; in other words, where the vertical and horizontal cloud growths are simply proportional, which again occurs where $h^2 = 2\sigma_z^2$, the result is

$$\chi_{max} = \frac{2Q}{\pi h^2 e u} \frac{\sigma_z}{\sigma_y} \tag{9}$$

Cloud Width and Height Formulas

Practical computations with dispersion formulas are often expedited by the construction of concentration isopleths. To do this, it is convenient to know the lateral distance, z_0 or y_0, where the concentration has dropped to p, percentage, of its value on the plume axis. For the Gaussian model the following formulas are an obvious application of Eq. 1:

$$y_0 = \left(2\sigma_y^2 \ln \frac{100}{p}\right)^{1/2} \tag{10}$$

and

$$z_0 = \left(2\sigma_z^2 \ln \frac{100}{p}\right)^{1/2} \tag{11}$$

Deposition and Washout

The question of what occurs at the boundary of interaction between an aerosol cloud and the underlying ground surface is clearly a very complicated one. Material is deposited on surfaces by a variety of physical processes, few of which are at all well understood. Gravitational settling, adsorption, particle interception, molecular and turbulent diffusion, and various chemical and electrostatic effects no doubt must be considered. A widely used treatment by Chamberlain[2,9] bypasses these effects and simplifies the problem neatly by defining a deposition velocity, V_g, as

$$V_g = \frac{\text{amount deposited/cm}^2 \text{ of horizontal surface/sec}}{\text{volumetric concentration above this surface}}$$

This definition can obviously be expressed in the following way:

$$V_g = \frac{\text{total deposition/cm}^2 \text{ of horizontal surface}}{\text{dosage (time integral of vol. conc.)/cm}^3}$$

Some values of V_g have been compiled by Chamberlain.[9]

For a continuous ground-level source, Chamberlain's deposition correction is obtained by multiplying the ground-level concentration (i.e., Eq. 1 with $z = 0$) by the deposition velocity V_g so that

$$\omega(x,y) = V_g \chi(x,y,0) \tag{13}$$

To account for the cloud depletion resulting from the deposition, the constant source strength Q is replaced by a quantity Q_x, depending on x, which is defined so that

$$\frac{\partial Q_x}{\partial x} = -\int_{-\infty}^{\infty} \omega(x,y)\, dy \tag{14}$$

i.e., Q_x is the total cloud depletion out to a distance x. Because σ_y and σ_z are functions of x, explicit relations for the dispersion (such as Sutton's) must be used to derive specific deposition formulas. When sufficient experimental data on σ_y and σ_z are available, a numerical evaluation can be attempted.

To account for washout (i.e., the removal of cloud material by rainfall), Chamberlain assumes that

$$Q_r = Q \exp\left(-\frac{\Lambda x}{u}\right) \tag{15}$$

where Q_r is a source-strength correction and Λ is the washout per second. Since rain removes material from the whole cloud depth, the process can be likened to radioactive decay because the entire cloud is affected uniformly rather than preferentially near the ground, and the shape of the cloud-distribution function is not altered. In this case the concentration from a ground-level source becomes

$$\chi = \frac{Q}{\pi u \sigma_y \sigma_z} \exp\left(-\frac{\Lambda x}{u}\right) \exp\left[-\left(\frac{y^2}{2\sigma_y^2} + \frac{z^2}{2\sigma_z^2}\right)\right] \tag{16}$$

where z is the vertical distance in meters. The amount of radioactivity deposited by rainout per second, ω_r, is obtained by multiplying Λ as in Eq. 16 and integrating with respect to z, as follows:

$$\omega_r(x,y) = \frac{\Lambda e^{-\Lambda x/u} Q}{(2\pi)^{1/2} u \sigma_y} \exp\left(-\frac{y^2}{2\sigma_y^2}\right) \tag{17}$$

Values of Λ, the washout per second, are the subject of considerable speculation, but very little observational information is available. However, if Λ is regarded as a continuously

varying parameter, Eq. 17 can be maximized with respect to it. The result is

$$\frac{\omega_r(\max)(x)}{Q} = \frac{1}{xe(2\pi)^{1/2}\sigma_y} \tag{18}$$

which gives the maximum values of washout by rain at any distance, x. The corresponding result for deposition depends on the form of $\sigma_z(x)$. As a rule of thumb, the maximum rainout should be roughly an order of magnitude greater than the maximum deposition, i.e., the suggestion is that

$$\omega_{\max}(x) \approx \frac{1}{10}\,\omega_{r\,(\max)}(x) \tag{19}$$

Radioactive-Cloud Dosage Calculations

Methods for computing the dosage to people as a result of exposure to a dispersing, radioactive, fission-product cloud are discussed[2] at length in USAEC Report AECU-3066, Chapter 8. Two problems were considered: (1) internal exposures due to inhalation of fission products and (2) external whole-body exposures. The internal dosage and the external beta dosage are essentially proportional to the local concentration at the point of reception, i.e., to χ of Eq. 1. Accordingly, for internal dosage and external beta dosage, relations given in terms of Sutton's equations can be converted into the form of the generalized dispersion model by use of the following substitutions:

$$\sigma_y^2 = \frac{1}{2}\,C_y^2 x^{2-n} \tag{20}$$

and

$$\sigma_z^2 = \frac{1}{2}\,C_z^2 x^{2-n} \tag{21}$$

External gamma dosage from an airborne cloud can be conveniently calculated from nomograms prepared by Holland.[2,10] Although these graphs were prepared on the basis of Sutton's dispersion model, they can each be entered on the scale labeled "σ," and therefore they are immediately adaptable to the generalized dispersion formulation. The value to be employed on the σ scale of Holland's graphs is, simply, $\sigma = (\sigma_y \sigma_z)$. (F. A. Gifford, Jr.)

Treatment of Low-Activity-Level Waste Water

Nuclear waste disposal has been principally concerned with the treatment, storage, and ultimate disposal of highly radioactive reactor fuel-processing waste. However, the volume of lower activity waste arising from laboratory facilities and chemical processing is orders of magnitude greater. Thus, means for decontaminating such waste at a low cost per unit volume are essential.

Although many papers discuss the design and operation of various plants for decontaminating low-level waste, there is a distinct need for a comprehensive summary and evaluation of the available data. Such information would be valuable to those having the responsibility for selecting and designing low-level waste-treatment plants; it would further serve as a guide for expenditures of time and funds for research in an effort to present a more complete picture of these treatment schemes. Obviously, such a short review as this cannot completely fulfill this function, but it is hoped that it will serve as an initial step in the more comprehensive treatment of the problem.

Salient facts pertaining to six waste-treatment plants located in Europe and in the United States are listed in Table V-1. In addition, other pertinent information about each facility is discussed in the following paragraphs.

Harwell, England

Two waste-water treatment plants are in operation at Harwell,[11–14] and a third plant for treating high-level waste is under construction. The low-level plant utilizes a single-stage process consisting of phosphate precipitation for gross decontamination. Sufficient Na_3PO_4 is added to maintain a phosphate residue of 100 ppm in the plant effluent. Sludge containing 3 per cent solids is further processed by pressure filtration (to 16 per cent solids) or freezing, thawing, and vacuum filtration (up to 32 per cent solid) and is finally put into drums and buried at sea.

The intermediate-level plant utilizes three stages. The first stage consists of phosphate precipitation and the addition of a conditioning agent for efficient coagulation. Sufficient calcium and phosphate are added to obtain a PO_4^{-3}

Table V-1 CHARACTERISTICS OF LOW-ACTIVITY-LEVEL WASTE-TREATMENT FACILITIES

Installation	Activity designation of waste stream	Activity in waste, μc/ml	Volume of waste, gal/day	Treatment units	Chemicals employed	Plant efficiency, % cumulation			Sludge treatment and disposal	Estimated operating cost (per 1000 gal)
						Alpha	Beta-gamma	Others		
Harwell, England	Low level	Not available	300,000	Pretreatment batching tanks, chemical dosing tanks, clarifiers, and posttreatment tanks	NaOH and Na_3PO_4	95	75		Pressure filtration or freezing, thawing, and vacuum filtration; burial at sea	£1.2[a,b] (~ $3.35)
	Intermediate level	Alpha, 2.7×10^{-5}	12,000–24,000	3 stages: (1) mixer and pulsator or precipitator, (2) mixer and precipitator, (3) vermiculite columns	Stage 1: NaOH, $CaCl_2$, or $Ca(NO_3)_2$, and Na_3PO_4 Stage 2: $FeSO_4$ and Na_2S Stage 3: None	Stage 1: 99 Stage 2: 99.8 Stage 3: 99.95	89 91 99.5–99.8		Pressure filtration or freezing, thawing, and vacuum filtration; burial at sea	£1.2[a,b] (~ $3.35)
Marcoule, France	Medium activity	0.02–1.5	40,000–53,000	Mixer and settler	$Ca(OH)_2$ and NaH_2PO_4, $Al_2(SO_4)_3$, tannin, and Separan	[c]	[c]		Vacuum filtration; burial on land	9500 francs[b,d] (~ $2.25)
Mol, Belgium	Cold	10^{-5}	117,000[e]	2 stages: (1) mixer and clarifier, (2) trickling filter	Stage 1: Na_3PO_4 and $Ca(OH)_2$ Stage 2: None	Total:[f] 56–89	30–89		Sludge thickener and sand drying beds	Not available
	Cool	10^{-3}	32,000[e]	4 stages: (1) pretreatment tanks, (2 and 3) mixer and accelerator, (4) filter or adsorber bed	Stage 1:[g] NaOH or $CuSO_4$, and $Na_2S_2O_3$ Stage 2: $Fe(SO_4)_3$ or $BaCl_2$ and $Fe_2(SO_4)$; *p*H adjusted Stage 3: $Ca(OH)_2$	Total:[h] 98.9	85.2	Sr: 94.5	Sludge thickener and sand drying beds	Not available

					Stage 3: $Ca(OH)_2$ and Na_3PO_4; *p*H adjusted Stage 4: None					
Los Alamos Scientific Laboratory	Laboratory waste	5×10^{-3}	173,000[e]	2 stages: (1) mixer, influent-holding tanks, flocculators, and sedimentation basins, (2) sand filter	Stage 1: NaOH, $Ca(OH)_2$ and $Fe_2(SO_4)_3$, coagulating agent Stage 2: Calgon	Total:[i] 99	Not available		Sludge thickener, vacuum filtration; burial on land	$5.86[i,b]
Idaho Chemical Processing Plant	Low activity	Not available	2,000[j]	Evaporator	None		99.996		Storage in tanks	Not available
Oak Ridge National Laboratory	Process waste	5×10^{-4}	750,000[k]	Mixer, flocculator, and settling basins	$Ca(OH)_2$, Na_2CO_3, and clay	90	88	Sr: 84 Cs: 86 TRE: 86 Pm: 94 Co: 78 Ru: 76	Disposal to seepage pit	$0.40[b]

[a]Cost increased by approximately 6 per cent for freezing treatment of sludge.
[b]Cost includes estimated charge for plant amortization.
[c]Incomplete experience with new facility.
[d]Based on treatment of inactive liquid.
[e]Plant capacity.
[f]Total for first six months; beta-gamma activity is dependent on waste composition.
[g]Probable chemicals to be used; based on laboratory tests and waste composition.
[h]Total for first four months; does not include treatment by adsorber.
[i]Based on operational experience during 1953 for treatment of 6.9 million gal of waste; plant is still in use.
[j]For operating period Nov. 11, 1954, to Mar. 18, 1955, at 30 per cent of plant capacity.
[k]About 430,000 gal of waste per day is treated.

to Ca^{++} ratio of 1.6, with a minimum dosage of 50 ppm Ca^{++}, and 40 ppm Fe^{++} or Fe^{+3} is added for coagulation. Most of the strontium is removed in this stage. The second stage employs sulfide precipitation for ruthenium removal (20 ppm Fe^{++} and S^{--}), and a conditioning agent is added for coagulation (20 ppm Ca^{++}). The effluent is passed through vermiculite columns for cesium removal and for a final strontium cleanup. The columns also serve as efficient filters for removing solids carried over from the precipitation processes. The final effluent from both plants is released into the Thames River, which serves as London's main source of drinking water.

Actual operating costs are not given, and the estimated costs given in Table V-1 are based on values listed by Burns and Glueckauf[12] for alternative flow sheets. The high-level plant now under construction[11] will include chemical precipitation, ion exchange, and evaporation to provide more flexibility and greater treatment efficiency.

Marcoule, France

Although operating data are not available for the plant at Marcoule, information[15,16] pertaining to the plant is of interest because of the level of activity to be handled by single-stage chemical precipitation. Primary treatment will consist of phosphate precipitation and the addition of tannin for increased efficiency. Alum and Separan, a commercial product of the Dow Chemical Company, will probably be used as conditioning agents for coagulation. Sludge containing about 2 per cent solids will be processed by vacuum filtration (to an expected 30 to 40 per cent solids), put into drums, and buried on land. A volume reduction to about 1 to 2 per cent of the waste treated is expected. Effluent from the treatment plant will be discharged into the Rhone River.

Mol, Belgium

Two treatment plants are in operation at Mol, and a third plant for "warm" (more active) effluents is under consideration.[17] The "cold" plant provides essentially a two-stage treatment, with the first stage using phosphate precipitation and the second stage using a trickling filter. Gross decontamination efficiencies vary because of the variable radionuclide composition of the waste. Although the filter reduces the peak concentrations of radionuclides, a continuous release of activity results, and its regular use is questionable.

The "cool" plant utilizes four stages. The first stage is for the removal of radioactive iodine; the second is for the removal of uranium and radioactive sulfur and tellurium by a $BaSO_4$ precipitation; the third is principally for the removal of strontium by phosphate precipitation; and the fourth is an ion-exchange stage using lignite as the sorptive material. In both plants the sludge is dried on sand beds. The effluent from the treatment plants is discharged into a small river near the plant. Present considerations for treating "warm" waste include the use of ion exchange and evaporation.

Los Alamos Scientific Laboratory

The major radioactive isotope that contaminates laboratory waste at Los Alamos is plutonium.[18,19] A two-stage plant utilizing ferric hydroxide precipitation followed by sand filtration is employed for removal of plutonium from laboratory wastes. The sand bed acts as an effective filter for solids carried over from the precipitation process, although various coagulating agents are used to reduce the amount of solids reaching the filters. Sludge containing 9.2 per cent solids is treated by vacuum filtration (to 27.5 per cent solids), put into drums, and buried on land. The amount of sludge remaining after treatment is about 0.2 per cent of the volume of waste treated. The effluent is discharged into a nearby canyon, where the liquid completely disappears by eventual seepage into the ground.

Idaho Chemical Processing Plant

At the Idaho Chemical Processing Plant,[20–22] process water contaminated with small quantities of radionuclides is evaporated; the condensate is discharged by an injection well to the water table; and the concentrate is stored in tanks. The evaporation technique is used because of limitations on the discharge of activity to the environment. Operating costs of such treatment facilities are higher than those for the other processes described in Table V-1, but no value is presented for comparison because the available cost data[20] are based on the treatment of a small volume of liquid. During the five-month period of operation for which cost information is available,[20] the waste handled

utilized only about 30 per cent of the capacity of the system.

Oak Ridge National Laboratory

The waste-water treatment plant at the Oak Ridge National Laboratory[23] utilizes single-stage calcium carbonate precipitation, primarily designed for the removal of strontium and the rare earths. Clay is added for removing cesium and for improving decontamination from cobalt and ruthenium. Untreated sludge, amounting to 0.3 per cent of the volume of waste treated, is transferred to seepage pits. The effluent from the treatment plant is released into White Oak Creek.

Conclusions

As previously discussed, the treatment of waste water depends on the specific nature, concentration, and quantity of radioactive and stable materials involved; the quantity of waste; and the specific environment into which the effluent is discharged. Thus there is no single method of waste-water treatment. Chemical treatment can effect a removal of about 99 per cent of the alpha contamination and 95 per cent of the beta-gamma contamination when multistage equipment is employed. In general, single-stage treatment is at a disadvantage when more than one radionuclide is to be removed from solution, since the most efficient processes for individual radionuclides may not be compatible. Filtration of the effluent from a chemical precipitation step will improve the over-all efficiency by removing solids containing radionuclides. Ion exchange will further increase the efficiency of a plant employing chemical precipitation. The general conclusions on waste-water treatment are summarized in a statement by Burns and Glueckauf,[12] as follows: "Apart from total evaporation of the water, there is no single process that will permit the conversion of a dirty radioactive effluent to drinking water. A multistage process is thus required." (K. E. Cowser)

Strontium Movement in the Soil

The longer lived radionuclides of nuclear waste effluents and the fallout from the testing of nuclear devices eventually reach the soil and are subsequently redistributed by percolating water. Strontium-90 is of particular concern since it is a long-lived radionuclide and is quite hazardous because of its tendency to become incorporated in the bone. The movement of Sr^{90} in the soil is of interest because it affects the level of surface-water contamination and influences propagation through the food chain.

There are numerous factors that influence the movement of Sr^{90} in the soil:[24–27] (1) the surplus of water in the soil and its rate and direction of movement; (2) the cation-exchange capacity and nature of the clay minerals in the soil; (3) the presence of materials in the soil which will form relatively insoluble compounds with strontium; (4) the mass of material through which the strontium solution must percolate before reaching surface streams; (5) the chemical composition of the percolating water, including *p*H; (6) the occurrence of natural weathering and erosion; and (7) the presence of natural flora and fauna and the resulting biological uptake of strontium.

For a given site, most of these parameters remain essentially constant. Fluctuations in the surplus of water in the soil will occur periodically,[24] except where there is continuous release of large volumes of liquid nuclear wastes; but, over extended periods of time, even the natural fluctuations tend to average out, and the movement of strontium, as well as other radionuclides, can be considered to be a continuous process. From site to site, however, all the movement parameters vary over a considerable range, and thus the measurement of strontium movement at a particular site does not have general application. On the other hand, if conditions affecting the specificity of strontium sorption, and thus affecting strontium movement, are known, local site conditions can be improved to restrict the movement of Sr^{90}. Therefore a more detailed discussion of some of the parameters seems to be in order.

Effect of Cation-Exchange Capacity of Soil

The cation-exchange capacity of the material through which nuclear waste percolates is undoubtedly one of the important parameters in limiting Sr^{90} movement,[24,28] but its role is often overemphasized. With the possible exception of cases in which deionized water is used for cooling purposes, the quantity of radionuclides in the soil solution is several

orders of magnitude lower than that of the competing chemically equivalent stable elements. The sorption of strontium from solution by ion exchange is governed by the laws of mass action;[26,29] thus the magnitude of the selectivity coefficient for strontium, compared with the magnitudes of the selectivity coefficients for the stable elements of the solution, is as important as the total cation-exchange capacity of the sorbent. The magnitude of the selectivity coefficient is an especially important consideration when the ion-exchange complex in the soil is heterogeneous and contains small amounts of exchangers that have unusually high affinities for radiostrontium. It is also an important consideration when the selectivity can be appreciably altered by treatment of the waste stream.

Sorption Mechanisms

Numerous workers are attempting to determine the mechanism of strontium sorption by soils and minerals.[26,28–31] Cerrai et al.,[31] in comparing the sorptive properties of natural and modified bentonites for radiostrontium, found that strontium removal from distilled water increased as the *p*H was increased; but, at a very high *p*H, a reduction in strontium removal occurred because of increased ionic activity of the solution. This agrees with previous observations by other workers.[32,33] Cerrai et al. used 1 g of bentonite per liter of water to sorb strontium from distilled water at *p*H 7.5 and *p*H 9.5 and found that there was little effect when the nominal concentration of strontium was increased from the mass equivalent of 12.5 μc to 1.25 curies of Sr^{90} per liter. At strontium concentrations greater than 1.25 curies of Sr^{90} per liter, the percentage of strontium removal decreases. The effect seems to be that of simple mass action, since the concentration of strontium approaches the total capacity of the exchanger. At these higher concentrations the strontium ions compete among themselves for exchange sites on the sorber. Spitsyn et al.[28,29] also found that the process was controlled by simple mass action and that the level of radioactivity had little or no effect on the degree of radiostrontium sorption.

Because the sorption of strontium is controlled by mass action, the concentration and nature of the competing cations affect strontium removel. Thornthwaite et al.[24] state that the leaching efficiency of $CaCl_2$ is approximately ten times greater than that of NaCl. The leaching efficiencies of these salts are not influenced by *p*H, but they are influenced by the exchange capacity of the sorbent. On the other hand, the leaching effectiveness of water is markedly affected by *p*H because it exerts an influence on the ionic concentration of the leaching solution. Spitsyn and Gromov[29] found the following order of decreasing inhibition of strontium sorption:

$$Ba^{++} > Ca^{++} > Mg^{++} > H^{+} > NH_4^{+} > K^{+} > Na^{+}$$

Spitsyn and Gromov further suggested that the presence of CO_3^{--}, SO_4^{--}, or $C_2O_4^{--}$ in solution causes strontium to form relatively insoluble salts. This does not change the mechanism of the sorption reaction, however; it merely decreases the quantity of strontium available for ion exchange. Ames et al.[30] have further considered the removal of strontium from solution by a calcite-phosphate mechanism and have obtained rather high strontium removals by passing a waste solution containing PO_4^{-3} through a bed of calcite ($CaCO_3$).

The presence of insoluble calcium phosphates in calcareous soils and calcium-aluminum and aluminum phosphates in neutral and slightly acidic soils suggests that the phosphate reaction might be of considerable importance in effecting strontium retention. Frondel[25] has suggested that strontium might be incorporated into minerals of the alunite structure type, such as crandallite, at near neutral *p*H and that the phosphate reaction would occur readily in formations containing these minerals. Spitsyn et al.[28] found that minerals varied considerably in their ability to sorb strontium. Because the solutions contained little or no competing ions, the sorbents having the highest ion-exchange capacities sorbed strontium most efficiently. On the other hand, McHenry[33] reports that the effectiveness of strontium sorption by the various soils in the vicinity of Hanford is not a simple function of ion-exchange capacity.

Effect of Natural Weathering and Erosion

The occurrence of natural weathering and the erosion of sediments can cause considerable movement of strontium sorbed on surface soils. Menzel[34] found that the loss of Sr^{90} in fallout was related to the soil cover and slope on plots growing agricultural crops (Table V-2). The

Table V-2 Sr^{90} IN FALLOUT AND RUNOFF AS RELATED TO SOIL COVER AND SLOPE*

	Site 1†		Site 2‡	
	Sr^{90}, μμc/sq ft	Soil loss, tons/acre	Sr^{90}, μμc/sq ft	Soil loss, tons/acre
Fallout	257		180	
Runoff from plot covered with:				
Corn	11.5	>1	2.5	0.6
Oats	10.5	>1	0.81	0.3
Clover	0.9	0.03		
Peanuts			2.6	0.7

* Summarized from data of Menzel.[34]

† Sampling site: 72.5-ft-long plot with a 16 per cent slope, La Crosse, Wis. Sampling period: Mar. 13 to Aug. 18, 1957.

‡ Sampling site: 83-ft-long plot with a 3 per cent slope, Tifton, Ga. Sampling period: Mar. 11 to Dec. 2, 1957.

soil cover and the slope acted primarily in affecting the amount of erosion; thus it appears that much of the Sr^{90} was associated with the sediments. Infiltration of water would also be expected to vary with vegetative cover; consequently, the relation is not simple.[35]

Methods for Limiting Strontium Movement

Spitsyn et al.[28] observed that radionuclides, especially radiostrontium and radioruthenium, moved much farther in the soil than laboratory saturation studies predicted. Two plausible explanations are that (1) the efficiency of the solid-solution contact was lower in the field because of channeling and larger effective particle size and/or (2) the sorption front was rather diffuse and thus allowed a small percentage of the activity to be observed some distance beyond the average extent of movement for the particular radionuclide. The use of the saturation capacity for predicting the extent of movement in a continuous ion-exchange system is essentially equivalent to assuming a sharp dividing line between saturated and unsaturated regions.[27] The saturation capacity can be used more realistically to estimate the average movement of the radionuclide. However, in the movement of radionuclides, especially Sr^{90}, fractions of the activity which are much smaller than the saturation quantity are usually of concern. The curvilinear nature of the moving sorption front has been discussed in some detail (references 24, 27, and 36 to 38), but it suffices to state that the concentration of activity remaining in solution decreases as the effective length of travel of the percolating solution increases. The extent of the sorption front is dependent, among other things, on (1) the selectivity coefficient for the radionuclide compared with the stable ions of the solution, (2) the flow rate of the solution, (3) the total concentration of stable salts, (4) the kinetics of the reactions, and (5) the particle size of the medium.[39]

At first glance it would seem improbable to be able to increase the mass of soil through which the strontium must percolate before reaching surface streams. However, if the radioactive contaminant is released to the surface (i.e., by fallout or accidental release of fission products), the effective path through the soil can be lengthened by promoting infiltration of the solution. Much more efficient contact is made as the solution percolates through the soil mass than when it moves over the surface. Increased infiltration can be obtained by selective use of cover crops or by terracing. Such practices have been recommended for reactor site improvement,[40] although additional work is necessary for a more complete evaluation.

The movement of strontium in the soil involves an interplay of several factors, and, by judicious modification of local environments, the movement of Sr^{90} in the soil can be limited. Because of the hazard of this isotope, however, its migration from waste-disposal sites must be closely monitored. (D. G. Jacobs)

Biological Uptake of Radionuclides

Radionuclides reaching the environment from nuclear waste effluents or from the testing of nuclear devices can create hazards for man, animals, and plants. These hazards, which originate from external radiation or from the internal radiation from assimilated radionuclides, can upset the balance of the ecological system. As a result a new equilibrium may be established at lower efficiency. To understand and predict the effects of radionuclides released to the environment, several ecological programs, tracing pathways and rates of turnover of the radioisotopes through biota, water, soil, and air, are under way. In addition, much work is being conducted on the biological uptake and translocation of radionuclides in segments of

the environmental cycles, especially in those leading to man.

Uptake of Radionuclides from Fallout on Land

Widespread radioactive contamination of terrestrial environments originates almost exclusively from world-wide or close-in fallout from weapon tests. The biological consequences of close-in fallout at various distances from Ground Zero have been discussed by Lindberg.[41] The biological consequences are a function of the physical and chemical characteristics of the contaminants. Particulate debris formed during a nuclear explosion is principally spherical, and it picks up radionuclides from the fission cloud. When devices are detonated at high altitudes, the particles are smaller than those of a detonation that takes place near the ground; also they travel farther, and, in general, the radionuclides they pick up are more soluble. Biological availability increases with solubility. Autoradiographs of plant leaves collected in the close-in fallout area show that most of the external contamination is due to particles less than 44 μ in diameter and that the fraction of the total contamination associated with particles less than 44 μ increases at increasing distances up to more than 100 miles from Ground Zero. Animals feeding on plants ingest and accumulate physiologically important long-lived isotopes. Thus, six months after an explosion, a "hot spot" was found 132 miles from Ground Zero, and the radioactivity in the bones of local jack rabbits was the maximum found at any point. This activity was mainly due to Sr^{89}.

The uptake of radionuclides by plants through the leaves and the consequences for animals feeding on these plants have also been studied by Morgan.[42] He compared the concentration of various radioisotopes in rye grass, grown for 30 days in the open on a nutrient solution in expanded vermiculite, with the concentration of radioisotopes in rain water collected during the same period. Since the radionuclides, with the exception of Sr^{90}, are ordinarily only slightly taken up by and translocated in roots, it was assumed that activity in the plants was mainly the result of leaf uptake. The data indicated that the uptake of Cs^{137} (24 to 29 per cent of deposited activity) is slightly less than the uptake of Sr^{89}-Sr^{90}. The uptake of Zn^{95} is higher than that of Sr^{90}, and the uptake of Ce^{144} is still greater. There was also an indication that, with increasing leaf area, an increasing quantity of radionuclides per unit surface area was taken up. The amount of the various isotopes taken up seems to be determined by the initial rate of sorption on the leaf and the rate at which the isotopes wash from the leaf. These data clearly indicate that plants growing out doors will contain the entire spectrum of radionuclides present in the current fallout. The data further indicate that the estimation of plant contamination, based on the relative rates of root uptake for various radionuclides, is not valid. Based on various assumptions, Morgan[42] calculated that the permissible dose for a cow is 1 rad/day and that the maximum uptake from fallout to date is 10^{-3} times this value.

Uptake of Radionuclides from Nuclear Waste Effluents

The release of radionuclides from reactor effluents differs from fallout from atomic weapons in that the activity is not so widely dispersed but, rather, is concentrated primarily in local environments. The release of such effluents into the Columbia River is discussed in a Hanford report.[43] As in bomb fallout, there is rapid decay of total activity shortly after release. After 6 hr, less than 20 per cent of the activity initially released remains. Therefore the discussion[43] is focused primarily on the more persistent radionuclides P^{32} and Zn^{65}. The concentration of radionuclides in the river water varies inversely with the flow rate; consequently the uptake by men and animals who drink the river water or by plants that grow in, or are irrigated with, river water varies inversely with flow rate. The uptake of radionuclides by produce irrigated with river water is often below the detection limits. Zinc-65 is prominent among the biologically important radionuclides originating from river water which has been found in measurable quantities, especially in cereals and alfalfa.

Uptake from Fallout on Water

The accumulation of radioisotopes from fallout by salt-water algae has been discussed by Akamsin et al.[44] who have studied the uptake of radionuclides by algae to find a suitable indicator organism for the detection of fallout on

water surfaces. As a result of diffusion and water movement, activity from fallout is rapidly dispersed and diluted to low levels in a medium with a high background of natural K^{40}. Plankton, which has a large surface area and a high mineral content, makes a good indicator, but it is difficult to collect. Therefore, as an alternative, brown algae were assayed. Their activity, as that of plankton, ranges from three to six times the activity of the natural K^{40}. Calculations based on the ratio of K^{39} to K^{40} and radiospectrograms of the brown algae indicated the presence of Sr^{90} and Ce^{144}; whereas, in green algae, only a slight accumulation of Sr^{90} was observed. A wider use of brown algae for the detection of radioisotopes in sea water was therefore suggested. From a study on gross beta activity of algae at Eniwetok,[45] it appears that the highest beta activity in the algae occurs at the time and place of highest fallout. Decline of activity in the algae was faster than could be accounted for on the basis of physical decay and appeared to be due to leaching by relatively uncontaminated currents. Later observations showed, however, a slower loss of activity in the algae than in the water, indicating uptake and accumulation of long-lived radioisotopes. In general, however, there was a close relation between the isotopes in algae, water, and sand. Fleshy algae appeared to contain a higher concentration of radionuclides than coralline species.

Uptake by Fish and Water Fowl[43]

Because of the high mineral concentration in salt water and in salt-water algae, the relative accumulation of radionuclides is low compared with that of fresh-water algae. In the Columbia River there is a marked concentration of radioisotopes in the fish (suckers) that feed directly on the algae. Fluctuations in flow rate of the river and in temperature cause changes in the radionuclide concentration and metabolic activity of the algae and the fish. These changes influence directly the radioactivity of biota feeding primarily on algae and detritus and less directly the activity of omnivorous and predatory fish and of local water fowl. Least affected are the migratory fish and water fowl. The hazard from human consumption of the fish is greatly reduced because more than 90 per cent of the activity is due to P^{32}, which is predominantly concentrated in bones and scales. The permissible annual consumption of white fish taken a short distance downstream of the Hanford area was calculated to be 50 lb per person. The concentration of P^{32} in ducks feeding in this area is of the same magnitude as that in fish. During the fall hunting season, however, the autochthonous population is diluted 200- to 1000-fold by migrating uncontaminated ducks.

Uptake by Oceanic Biota

The uptake of the radionuclides from reactor effluents by oceanic biota was most apparent in oyster cultures in Willapa Bay, north of the mouth of the Columbia River. This marked uptake by a shellfish warrants special attention, and a short-term study by an oceanographic group on this subject was advised. As discussed in this review, much work must be done before integration of detail studies into a comprehensive picture of the cycling of radionuclides, matter, and energy in various ecological systems can be effected. (M. Witkamp)

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Section VI

CURRENT EVENTS

Westinghouse Testing Reactor Incident

During operation of the Westinghouse Testing Reactor (WTR) at Waltż Mills, Pa., on Apr. 3, 1960, a fuel-element failure occurred. The failure was accompanied by release of fission products to the primary coolant system and discharge of some gaseous fission products to the atmosphere. Since the radiation dose levels were, in general, not high, no serious personnel exposure occurred. A recent report by the staff of the WTR[1] describes the events that led up to the activity release and then discusses in considerable detail the problems faced and the methods that are being used to put the reactor back into operation. As a result of the incident, the AEC has restricted future operation at various specified power levels to certain minimum coolant-flow rates, which are intended to preclude any significant amount of boiling in the core, and has prescribed the severity of the boiling pattern as indicated by the boiling-detector noise level.[2] This Review is concerned primarily with the events that led to the incident and the causes of the incident.

Description of the Reactor

The WTR, a modified MTR type system,[3] is water cooled, water moderated, and beryllium reflected. The fuel elements are in the form of three concentric cylinders approximately 42 in. in over-all length. The outer cylinder is $2\frac{1}{2}$ in. in outside diameter; the fuel plates are 125 mils thick; and the coolant-flow gaps are nominally 94 mils wide. The fuel-bearing region is 36 in. long and consists of a sandwich type assembly having 36-mil-thick aluminum cladding and a 53-mil-thick "meat" section. The meat consists of a 13 per cent uranium-aluminum alloy, with the uranium fully enriched in U^{235}. Each element initially contains 200 g of fuel. The three cylinders of the fuel element are arranged coaxially around a central nonfuel-bearing "basket" or mandrel. This basket may also be used as a receptacle for samples.

The core structure consists of upper and lower hexagonal tube sheets connected by two $^{11}/_{16}$-in.-ID cylindrical shroud tubes in which the fuel elements are placed. The center-to-center distance between fuel elements is $3\frac{1}{8}$ in. The structure is a slightly unsymmetrical hexagon having 86 positions for fuel elements and control rods and 7 high-pressure thimbles for test loops. At the time of the incident, 69 positions were occupied by fuel elements and 9 positions were occupied by control rods. Each control rod consists of a cadmium-containing poison section followed by a standard fuel-element section.

Operation of WTR Prior to Incident

About three months prior to the incident, the Advisory Committee on Reactor Safeguards (ACRS) had agreed that it would be safe for the power of the WTR to be raised from 20 to 60 Mw, with the higher power level being approached through a planned, stepwise program. This power-increasing program was in effect at the time of the incident, and the reactor had been operated at power levels up to 45 Mw with "rated" coolant flow without difficulty for some time prior to the incident.

A number of low-coolant-flow boiling tests had been conducted in order to determine the upper limits of operation. On the basis of theoretical considerations and data obtained when operating at 20 Mw with 2500 gal/min core flow and at 40 Mw with 4700 gal/min core flow, the following relation between the core

flow and the power level at which boiling first occurs was formulated:

$$P = 0.0984\ F^{0.705}$$

A similar relation had been deduced at Oak Ridge National Laboratory (ORNL) for the Oak Ridge Research Reactor (ORR); i.e.,

$$P = 0.0244\ F^{0.8}$$

In both cases, P is power in megawatts, and F is coolant flow in gallons per minute; however, the flow of the ORR used in the formula is total flow, whereas that of the WTR is flow through the core only. In the tests the onset of boiling was detected by perturbations in the power level picked up by an ionization chamber connected to a Brush recorder. The magnitude of the oscillations observed on the Brush recorder was taken as an indication of the amount of boiling in the core. The procedure used was similar to that used in the ORR boiling tests performed[4] in 1958.

Operation of WTR at Time of Incident

The reactor startup prior to the incident began on April 2. The power level was gradually increased, and a level of 40 Mw was reached in the afternoon. The primary coolant flow was 15,000 gal/min, and two-thirds of the flow was directed through the core. The reactor was then maintained at a power level of 40 Mw, except for a reduction in power on April 3 because of test-loop trouble. This trouble was found to be inconsequential, and the reactor was returned to a power level of 40 Mw within 1 hr. Some 9 hr later, on April 3, the reactor power was reduced to 30 Mw in preparation for the scheduled minimum-flow test, and the primary coolant flow was gradually reduced to 5250 gal/min (3500 gal/min through the core). During this time the boiling detector did not show any perturbations which would lead the operators to suspect boiling.

The power level was then raised and allowed to settle at approximately 34 Mw, as measured by the nuclear power instrumentation. It was soon observed that the power level was falling, and the operator, under instructions of the shift supervisor to raise the power level to 40 Mw, manually withdrew three of the control rods by amounts equivalent to 2 per cent of their travel. The resulting reactivity increase, together with reactivity added by the automatic control system, increased the reactor power level to approximately 37 Mw. Just before the power level reached 37 Mw, the demineralized-water monitoring channel alarmed. This alarm was acknowledged, and almost immediately several other alarms indicated high radiation levels in the various monitored areas. A few minutes later the power demand setpoint was reduced, and there was an immediate manual reactor cutback. Four minutes later the reactor was manually scrammed.

The reactor instrumentation had indicated a sharp momentary setback in power at the time of the first alarm and a return to full power 4 min later. The initial reduction in power is believed by the WTR staff to have occurred as a result of a reactivity decrease caused by fuel-element meltdown and consequent blockage of the coolant channel. It was suspected at the time, and later confirmed, that fission products were being released from the coolant-circulation head tank, which is vented to the atmosphere, and that they were producing radiation levels in the plant areas that were sufficiently high to produce alarms. The plant was evacuated.

Local radiation levels near the 8-in. water lines below the head tank, which is located at an elevation of about 250 ft above the core mid-plane, were as high as 40 r/hr. At other portions of the plant, the radiation levels reached 100 mr/hr. Most of the released fission-product activity was gaseous, and much of this escaped harmlessly from the head-tank vent. Calculations indicated that the total activity released to the primary system was about 5000 curies, and the airborne activity released in the initial burst was 93 curies, followed by an additional 168 curies before the stack blower was shut off. This discharge was well within the permissible seven-day average discharge. It is believed that a maximum of 800 curies of radioactive gas, consisting primarily of xenon and krypton, was ultimately released from the head tank.

The design of the vent system which allowed uncontrolled release of activity directly to the atmosphere was a weak link in the containment provisions for this facility. Although the activity release indicated for the current incident was within the permissible levels, a more serious accident obviously could not have been handled properly. In a letter from the AEC, it

is stated that[5] "the venting system for the process water head tanks and the water surge tank should be modified or eliminated to protect against releases of fission products to the atmosphere."

An independent survey of the environmental activity after the incident was undertaken by Nuclear Science and Engineering Corporation (NSEC). The samples obtained showed no appreciable increase in activity levels above the background values obtained on previous surveys made by NSEC during the preceding three years. There is no indication of exactly when the survey was made, but either it was made after short-lived activity had decayed or the results indicate that the activity released was predominantly xenon and krypton.

Heat-Transfer Analysis

Calculations of the heat flux under pessimistic assumptions indicate that it was lower than that required for burnout by a factor of at least 1.6 if the heat flux were distributed uniformly over the heat-transfer surface of the fuel element.[6] Consequently it seems highly probable that the fuel element which failed contained an unbonded area of sufficient size to raise the local heat flux to burnout conditions. This conclusion is substantiated by the normal behavior of another element under apparently identical conditions of power density and coolant flow. Although the possibility is not ruled out, it appears improbably that the failure would have occurred with a sound element. Calculations made by the Westinghouse staff after the incident occurred showed that an unbonded area as big as 1 in. in diameter on one side of the meat in a fuel element would be sufficient to cause a 100 per cent increase in the heat flux across the cladding on the opposite side of the meat; an unbonded area $\frac{1}{2}$ in. in diameter could cause about a 60 per cent increase, and an unbonded area as small as $\frac{1}{8}$ in. in diameter could cause about a 10 per cent increase. The presence of an unbonded area in the element that failed could not, of course, be determined from subsequent examination because of the destruction of the element. However, the examination showed areas where the meat had melted out of the aluminum cladding and left void areas, and thus there was evidence of extremely high central temperatures. Ultrasonic examinations of the reserve supply of unused fuel elements revealed that a significant number of them had blisters, or voids, i.e., unbonded areas, large enough to raise doubts as to their adequacy for service under stringent operating conditions. Consequently the specifications have been modified to reject all elements showing defective areas larger than $\frac{1}{8}$ in. in diameter under ultrasonic test.

In some of the defective elements that were sectioned for further examination, the void was not between the meat and the aluminum cladding; thus a question is raised, in these cases, as to whether a poor bond was at fault. Information has been received[7] that the voids could have been caused by hydrogen inclusion in the alloy ingot and that such inclusions can be avoided either by vacuum casting or by cycling the melt above and below the melting temperature to remove the hydrogen.

Actions Taken Following Incident

As a consequence of this incident, the following corrective actions have been taken by the operator:[8]

1. The initiation of a rigid inspection program of all cold fuel elements now on hand and currently being manufactured .... Fuel with defects larger than an equivalent diameter of approximately one-eighth ($\frac{1}{8}$) in. will not be used. The presence of such defects will be determined by ultrasonic means or by any better means which may become available.
2. All members of the WTR Operations Department have been instructed on the hazards of fast negative reactivity changes. The WTR Operating Procedures P-107 have been revised to cover the operation of control rods subsequent to sudden changes in reactivity.
3. The 60-Mw power escalation program will be modified to limit the amount of boiling in the core to a value below that permitted by License No. TR-2. At no time in the escalation program or the early 60-Mw operating cycles will the boiling pattern be permitted to be more severe than the proven pattern of Figure 13, WTR-49, 8000 gpm case.
4. The head-tank vent system was provided with a motor-operated valve which will be manually actuated to close at a nominal radiation level in the tank and will automatically close in the event of a sudden large increase in effluent activity level.

Conclusions

1. It is likely that either or both of the following two factors played a major role in the overheating and subsequent melting of the fuel element: (*a*) defective metallurgical bonding of

the fuel element and (*b*) inadequate coolant flow to the fuel element.

2. The detection of boiling is not a suitable means for controlling a reactor, even for an experiment, because it is not possible to distinguish between general nucleate boiling (safe) and the local bulk boiling that leads rapidly to failure.

3. The existing head-tank vent system, which permitted fission-product gases to be discharged external to the containment vessel, was a serious design deficiency.

4. The incident does *not* indicate that the proposed WTR operation at 60 Mw would necessarily be unsafe even with elements containing unbonded areas or blisters larger than $\frac{1}{8}$ in. in diameter.

5. If the operator had shut down the reactor at the time of the first downward power drift, the activity released to the coolant would very likely have been less than that experienced, inasmuch as it appears that the first downward drift of power probably was an indication of bulk boiling. Conceivably, there is even a chance that an immediate shutdown might have prevented failure. This analysis illustrates a problem pertinent to the operation of all swimming-pool type reactors, namely, whether to provide a safety procedure empowering the operator to set back or scram the reactor immediately upon observance of any of a set of stated phenomena regardless of the test being run.

(R. B. Korsmeyer)

Radiation Incident at the ORNL Multicurie Fission-Product Pilot Plant

On Mar. 8, 1960, the overexposure of an individual occurred during decontamination of cell 11 of the Multicurie Fission-Product Pilot Plant at ORNL. The source of the exposure was Ce^{144}-Pr^{144}, which was present in a drain trough. During the estimated 10 min in which the person worked in the cell removing trash from the floor and the trough, he received a dose of approximately 5500 rads of soft beta radiation to the surface of his left hand. An as yet unpublished report, which was prepared by the ORNL investigating committee (F. R. Bruce, C. D. Cagle, and W. H. Jordan), describes the events leading up to the accident and makes recommendations to prevent the recurrence of such an accident.[9] The description of the incident and the findings and recommendations of the committee are summarized here.

The Multicurie Fission-Product Pilot Plant consists of a block of 15 cells containing the process equipment required for recovering fission products from fuel-processing waste solutions. Wet chemical operations are used for the separation of individual fission products from the mixture. Final processing consists of conversion of the fission products to a desired source by drying, powdering, pelletizing, and, finally, encapsulation.

The first decontamination of the cerium powder handling cells (cells 11 and 12) since startup of the Multicurie Fission-Product Pilot Plant began on Feb. 4, 1960. During the ensuing three weeks, calcining furnaces and other equipment were removed from cell 11. By March 2, chemical decontamination, consisting of solution rinsing and scrubbing, was under way. During the decontamination program, unusual difficulties associated with the handling of cerium and praseodymium, which emit soft beta radiation, were encountered for the first time. For example, when $\frac{1}{4}$-in.-thick lead sheet presumed to be uncontaminated was removed, the underside was found to read greater than 50 rads/hr.

Efforts to decontaminate cell 11 were hampered by a plugged drain in cell 13, which communicates with a drain trough that is common to cells 10 to 13. It was planned to open the drain in cell 13 on March 4 and to clean the drain trough after the water had drained from it, but the work schedule did not permit unplugging the cell 13 drain until the 4 to 12 p.m. shift on March 7. The drain was unplugged, apparently without incident.

Description of the Incident in Cell 11

Eight people (one supervisor and seven operators) were working in the Multicurie Fission-Product Pilot Plant on the night of the incident. Between 12 midnight and 12:45 a.m. on March 8, the top of cell 11 was decontaminated, and at approximately 12:50 a.m. the cover block was removed. Between about 12:50 a.m. and 12:55 a.m., one of the operators equipped himself for cell entry. The supervisor in charge visually surveyed the work to be done and saw only welding rod and slag which had accumulated on

the cell floor. He then verbally instructed the operator to enter the cell and remove the welding rod and slag. Since the supervisor was not aware that there was also trash present in the drain trough on the north side of the cell, he presumed that the refuse could easily be removed in less than the allowable 5 to 6 min working time in the cell, and therefore the operator's residence time in the cell was not timed.

The operator entered the cell, picked up the welding rod and slag with a gloved hand, and placed it in a bucket. This debris filled the bucket to about three-fourths full. He then observed slag in the cell drain trough, which he scooped up and also placed in the bucket. At times he used small pieces of metal as a scoop for removing the debris. He did not use his right hand in order to keep it clean and available for climbing the ladder upon leaving the cell. Although no record was kept of the time he spent in the cell, the operator estimated it to be about 10 min. Between the time the operator entered and left the cell, the supervisor was working with other operators at cell 13. When the operator left cell 11, at about 1:05 a.m., carrying the bucket in his left hand at approximately thigh height, the space monitor located over cell 11 alarmed. The supervisor came over from cell 13 and placed a 20-gal "hot" disposal can in position to receive the bucket the employee carried. The radiation level 6 in. over the bucket that the employee removed from the cell was found to be 200 rads/hr. A later observation that the radiation level was 1 rad/hr through the bucket wall indicated the soft nature of radiation. The operator's dosimeters, which were capable of reading only up to 200 mr, were examined and found to be off-scale. It was learned that the operator had not read his dosimeter while he was in the cell.

Analysis of the Incident

A factor of paramount importance in explaining the high, unanticipated radiation background which resulted in this exposure is the common drain trough for cells 10 to 13. When the last official health physics survey of cell 11 prior to the incident was made on March 4, the trough was filled with water, which served as shielding for the soft radiation emanating from the Ce^{144}-Pr^{144} in the trough. The radiation field was 5 to 20 rads/hr approximately 5 ft from the cell floor. The supervisor was under the impression that the background in cell 11 ranged from 1 rad/hr to 200 rads/hr, with the latter level being close to some highly contaminated spots. It was expected that the work performed by the operator would be carried out in a field averaging about 1 rad/hr.

It is believed that the water shielding was drained from the cell 11 trough during the 4 to 12 p.m. shift on March 7, as a result of the common drain in cell 13 being unplugged. It is also possible that decontamination operations carried out in cells 12 and 13 during the period March 4 to 7 resulted in sizable quantities of radioactivity being flushed from these cells, via the trough, to cell 11.

The first survey of cell 11 following the incident was made by the shift health physicist at about 4:30 a.m. on March 8. He estimated the general field at 100 ± 10 rads/hr. At 2 ft from the edge of the trough, however, a high-range probe went off-scale at 200 rads/hr. The shift health physicist estimated that the operator's body was in a 100 rads/hr field while cleaning debris from the trough.

Development of the operator's film badge showed approximately 2.5 rems of penetrating dose to a depth in tissue equivalent to 1000 mg/cm^2; about 40 rems of moderately penetrating dose to a depth in tissue equivalent to 130 mg/cm^2; a skin dose through the coveralls (30 mg/cm^2 absorption) of about 80 rems; and a direct skin dose to a depth in tissue equivalent to 7 mg/cm^2 of about 100 rems.

Since radioactive material was handled by the employee, special attention was given to the exposure to his hand. This was estimated by exposing films buried to various depths in a phantom paraffin hand to debris taken from cell 12. The quantity of debris in a bucket identical to that used by the operator was adjusted to agree with the radiation level of the incident. From this experiment, the following hand exposures were estimated:

	Dose, rads
Surface of the hand	5500
Epidermal layer at depth of 1 mm	3200
Epidermal layer at depth of 2 mm	2200

On Mar. 18, 1960, symptoms of a mild beta-radiation burn appeared on the tip of the operator's left thumb. On March 23 a similar,

but milder, reaction became apparent on the third finger near the tip. By April 1 the skin began to separate from the tip of the thumb and the third and fourth digits, but by April 7 peeling had ceased and healthy new skin appeared.

Although the exposure from the incident was believed to involve only cerium-praseodymium, Ce^{137} could not be positively ruled out. After making various analyses, however, it was concluded that no significant cerium body burden was present and that the employee did not sustain significant inhalation exposure during the incident.

Conclusions and Recommendations

A number of factors contributed to the occurrence of this incident, the most important being the nature of the radiation involved. The soft beta radiation from Ce^{144}-Pr^{144} is difficult to detect and was encountered here for the first time in large quantities in a nonstandard operation. In the drain trough the beta radiation was completely shielded by the water present, and the radiation level increased markedly when the water shielding was drained. Other causes of the incident were, however, attributable to mistakes that were made.

The direct causes of the incident were the following:

1. The supervisor failed to make a radiation survey of the cell before it was entered, and the presence of a high radiation field thereby went undetected.

2. General, rather than specific, instructions were given for the work performed with the result that the operator, on his own initiative, expanded the scope of his work.

3. The employee departed from normal practice in failing to read his dosimeter. Had he done so, he would have become aware of the high-radiation field and left the cell at that time.

Indirect causes of the incident were the following:

1. The supervisor was responsible for more operators than he could adequately supervise in view of the hazardous nature of the work involved.

2. The operator was given inadequate job training, as evidenced by his action in picking up, with his hands, material that he should have suspected as being radioactive.

A further factor was the difference in the health physics procedures applied to maintenance personnel and to operators. Areas to be entered by maintenance personnel must be monitored by health physics, whereas health physics monitoring is not required for entry by an operator.

The investigating committee has recommended the following actions:

1. Definite operating procedures should be drawn up and employed for nonstandard operations, such as the one involved in this incident, as well as for standard operations.

2. An entry permit form should be filled out prior to entry into a high-radiation zone.

3. Full-time health physics coverage should be provided on all shifts during periods of high-activity-level nonstandard operation.

4. During periods of nonstandard operation involving high radiation levels, one supervisor should be responsible for no more than four operators. If supervision of more than four is desired, full cognizance of higher supervision should be assured, and their approval should be required.

5. Adequate signs should be posted to indicate the presence of potential hazards.

6. The use of integrated-dose screamers should be considered for controlling personnel exposure in the case of work performed in high-radiation fields. (C. D. Cagle)

Operating Experience with the OMRE

The Organic-Moderated Reactor Experiment (OMRE) was first brought to criticality on Sept. 17, 1957. Three weeks later, a mechanical failure occurred in a flow-deflector skirt located between the top and bottom grid plates. The core was unloaded and drained; a skirt of improved design was manufactured and installed; the core was reassembled; and criticality was again achieved 11 days after discovery of the failure.[10,11] Low-power physics tests were performed during the next four months. The control rods were calibrated, and flux traverses were made of a single element. Mass, void, flow, pressure, and temperature coefficients were measured.

On Feb. 1, 1958, sustained operation at power was begun. Reactor control tests at various power levels were run during February to study the behavior of the reactor under transient conditions.[11] Operation with core 1 was con-

tinued until late November 1958, when the system was shut down for cleanup, and the second core was installed. During its life in the OMRE, core 1 was in operation for 5600 hr, of which 4800 hr was at power. It generated 958 Mwd of heat, and at the time of removal the reactivity remaining was enough for about two additional months of operation. Core 2 was identical to core 1 except for the location of some of the fuel elements that were instrumented with thermocouples. Criticality with the second core was achieved on May 9, 1959, and operation at power was resumed in June of that year.[12]

Experience with Core 1

There were two general modes of operation with the first core loading.[12] In one, the purification system was not used, and buildup of decomposition products was allowed; in the other, the purification system was operated, and the concentration of decomposition products was controlled. Operating experience with "high-boiler" concentrations of up to 41 per cent was obtained. (The term high boiler as used herein denotes the polymeric decomposition products that are less volatile than *p*-terphenyl.) The maximum fuel-element surface temperature was held at 750°F or below, and the coolant temperature was, in general, held at 600°F, although there were periods of operation with the coolant at 500 and 700°F. Reactor thermal power ranged up to 12 Mw, with most operation at about 6 Mw. The coolant velocity through the core was 14.7 ft/sec during the power runs.

Tests were made during the 10-month operating period with core 1 to check out the system at various conditions, to study radiolytic and pyrolytic decomposition rates, and to determine the effect of high-boiler concentration on heat transfer. The activity of the coolant was monitored, and radiation levels about the system were measured.

On June 23, 1958, the reactor was shut down, and fuel element 3 was removed for examination in a hot cell.[13] During this shutdown, two finned aluminum experimental elements, HT-1 and HB-1, were installed in the core. In addition, fuel element 30, one of the seven original elements instrumented with thermocouples, was removed from the center of the core to a peripheral position because its central thermocouple had failed. After startup, excessive temperatures were noted for fuel element 30. Following a six-day period of low-power testing, the reactor cap was removed, and it was found that element 30 had not been latched into position in the grid plates. It was inserted properly, and operation was resumed.

During an 8.5-Mw power run in October, fission-product activity was detected in the coolant, and on Oct. 23, 1958, a large amount of activity was released. This was found to be coming from experimental fuel element HB-1, and the element was removed. In the following run, temperatures in element HT-1 began to increase, and it was also removed. An undamaged fuel element, No. 33, was also removed at this time for hot-cell examination.

After resumption of operation, the fission-product activity in the coolant was still excessive, and the reactor was shut down in order to clean up the system. When the core was unloaded, fuel element 30 was found to be stuck. The center section of the element was distorted, and a split was observed in one cover plate. The element had to be removed with a hoist, and about 8 tons of force was required to pull it through the upper grid plate.

The coolant was transferred from the reactor and decontaminated by distillation in the purification system. Loose particulate matter was removed from the system by flushing with xylene, which was passed through temporary filters. Manual cleaning of the expansion and drain tanks was required for the removal of deposits which had probably been there from the time of installation. After cleanup of the system, the restored coolant was found to be sufficiently free of activity to permit installation of the second core. Significant details of the operation of core 1 are discussed below, including reactor physics, coolant decomposition, fuel-element behavior, radiation levels and activity, and component performance.

Reactor Physics.[10,11,14] The reactor became critical at 250°F with a loading of 20 fuel elements containing 16.4 kg of U^{235}. The calculated critical mass was 26 per cent less than this, and the difference was attributed to an underestimation of the fast leakage. For power operation, the core was loaded with 31 elements to give an excess reactivity of 7.8 per cent. This excess was estimated to provide 3.9 per cent to allow for 1800 Mwd of burnup and 3.9 per cent to overcome equilibrium xenon and

samarium poisoning at up to 16 Mw. No allowance was required for heatup, since, as discussed below, the reactivity was about the same at 700°F as it was at 250°F. With 31 fresh fuel elements, the reactor could still be shut down at any temperature, even though the two control-rod pairs of highest worth remained fully withdrawn. (There is a total of six control-rod pairs.)

The void and mass coefficients were determined at three different fuel-element positions (the center position was not included). The mass coefficient at 250°F ranged from 0.035 to 0.011 $(\Delta k/k)$ per fuel element. The void coefficient, for a void generated in a fuel element, ranged from -1.18×10^{-3} to -0.66×10^{-3} $(\Delta k/k)$/liter at 250°F and from -1.58×10^{-3} to -1.2×10^{-3} at 350°F; it was negative at all conditions tested.

The reactivity coefficient for uniform temperature change was positive at low temperatures but became negative in the range 435 to 485°F. It was about $+1.0 \times 10^{-4}$ $(\Delta k/k)/°F$ at 250°F and about -1.2×10^{-4} at 700°F. This difference between the characteristics of the OMRE and those of a pressurized-water reactor such as the SM-1, which has a negative temperature coefficient at all operating temperatures, is attributed to the large moderator gaps between OMRE fuel box assemblies and not to the nature of the moderator. The temperature coefficient is positive at low temperatures because the increase in thermal utilization with temperature is more important than the increase in neutron leakage. The coefficient becomes negative at higher temperatures because the change in neutron leakage becomes the more important. The integrated change in reactivity with temperature is such that the increase between 250 and 460°F is almost equal to the decrease from 460 to 700°F.

In tests of the reactor control system at power,[11] the reactor behaved well in response to transient disturbances. However, when the system was subjected to reactivity transients without either automatic or manual control, low-frequency power oscillations were found to exist. The period of the oscillations exceeded 10 min. The oscillations were divergent at temperatures below 600°F but convergent at temperatures above 600°F.

The oscillations are attributed to the difference in the temperature coefficient associated with the volume within the fuel-element boxes and that associated with the moderator between the fuel-element boxes. As shown experimentally, the former is negative, whereas the latter is positive. Coolant flow through the core is upward through the fuel elements and then downward through the spaces between the elements. Thus the moderator between the fuel elements is always hotter than that within the elements, and the temperature difference increases with increasing power. This causes the reactor to have a significant power coefficient of reactivity. The coefficient is positive at temperatures below 600°F and negative at temperatures above. The magnitude of the power coefficient is small, being about 3×10^{-5} $(\Delta k/k)$/Mw at 550°F with full flow, and this is not believed to present any hazard to the safety of the reactor. The oscillation has presented no problem in operation of the OMRE.

Two peculiarities, unexplained at the time of reporting,[15] were found regarding the distribution of fuel burnup. The burnup profiles indicated by a gamma scan of element 3 were practically flat and did not show the pronounced peaking expected near the edges, as found in element 33. In addition, element 33, which was irradiated in a mirror-image position relative to the position of element 3, had 144 per cent greater burnup, although it was exposed during only 75 per cent more megawatt-days of reactor operation.

Coolant Decomposition. The major uncertainty about the feasibility of organic-cooled reactors has been the effects of radiolytic and pyrolytic products on the operation of the system.[16] Aside from economic matters regarding purification and replacement of the coolant, there was the important technical question of whether the high-boiler decomposition products would interfere with heat transfer in the core and in the heat exchanger. The OMRE was specifically provided with two means of checking on this effect: (1) selected fuel plates were instrumented with three thermocouples each to measure surface temperatures of the hottest plate and two thermocouples each to measure the temperature of the coolant emerging from the channel next to the hottest plate; and (2) a bypass leg in parallel with the core was provided with an instrumented heater section for measuring the heat-transfer coefficient.

The heat-transfer coefficient, as measured in the bypass loop, decreased with increasing

high-boiler concentration, but the decrease was less than would be predicted from the change in physical properties of the coolant.[12,17] The coolant properties changed with the increase in high-boiler content, as expected; i.e., the density and viscosity increased with increasing high-boiler concentration, and the specific heat decreased. When normalized to a velocity of 14.6 ft/sec, the heat-transfer coefficient at 600°F decreased about 17 per cent as the high-boiler content increased from 10 to 40 per cent. The heat-transfer coefficient as a function of high-boiler concentration was the same whether the high-boiler content was being increased or decreased, and the change in heat-transfer properties appeared to be perfectly reversible. This was concluded to be confirmation of the absence of significant deposits on the heat-transfer surface. Heat-transfer measurements in the core indicated the same type of behavior as that observed in the bypass heater leg, and no significant insulation of the fuel-element surface by decomposition products was believed to have occurred (but note the later discussion of core 2).

The coolant damage rates[12,17] measured in the OMRE for the Santowax OM coolant agreed well with those previously measured in in-pile loops. The rates of coolant decomposition and consequent high-boiler formation decreased with increasing high-boiler concentration. At a high-boiler concentration of 10 wt.% the decomposition rate was 0.26 molecule of coolant per 100 ev of energy absorbed, and the high-boiler formation rate was 0.13 molecule (average molecular weight of 460) per 100 ev absorbed. The corresponding values at 30 per cent high-boiler concentration were only 0.15 and 0.07. These rates are based on an estimate that 7.8 per cent of the fission energy is deposited in the moderator.

Low-boiler materials and decomposition gases were also found in amounts equal to 2 to 3 wt.% of the decomposition products. The low-boiler concentration remained in the range of 0 to 1 per cent during reactor operation.

The high-boiler products contained large fractions of substituted polyphenyls and triphenylenes. The most important low-boiler products included benzene, toluene, ethylbenzene, *p*-ethyltoluene, *m*- and *p*-xylene, *n*-propylbenzene, and indane. The gaseous decomposition products were mostly hydrogen, with appreciable quantities of methane, ethane, ethene, and smaller quantities of other gases.

The pyrolytic decomposition was determined to be insignificant at 600°F, but at 700°F it was no longer negligible compared with the radiolytic decomposition at very low power levels. Experience in operation of the purification still at higher temperatures confirmed that pyrolitic decomposition becomes excessive at 800°F and above.[17,18]

Fuel-Element Behavior.[13,15] As described above, undamaged fuel elements were removed after 549 and 958 Mwd of operation of core 1 and were transferred to a hot cell for examination. They were transferred in shipping casks filled with the heat-transfer medium isopropyl diphenyl. The fuel element with the longer exposure had received an average uranium burnup of 3.9 at.% and a maximum burnup of 9 at.%. The examination of these elements indicated excellent radiation stability up to the highest exposure observed. The stainless-steel cladding was not found to be corroded, carburized, nitrided, or affected in any adverse manner by the coolant; slight pitting in one area was ascribed to a fabrication defect.

All plates of the element with the shorter exposure were covered with a thin black residue that was found to be brittle but adherent. (A similar coating on the outside of the fuel element was fairly soft but appeared to harden on drying when removed from the shipping cask.) Under magnification, small blisterlike blemishes were observed in the film, principally on the fueled portions of the plates. Optical measurements of sectioned portions indicated an average film thickness of 0.00025 in. and a maximum of 0.00037 in.

The plates of the undamaged element having the longer exposure were covered with a black residue of varying appearance. In some areas the film was thin and smooth, whereas in others there was a coarse thick residue. No relation between plate temperature and film thickness was discerned. Some residue resisted normal methods for removal. It was found that among various reagents tested, hot aqueous Turco 4502 solution was singularly effective. Also, satisfactory removal was possible if the elements were heated in 500°C air. Micrometer measurements of the film thickness showed a maximum thickness of about 0.006 in. and an average of about 0.002 in. Approximately 40

per cent of the material was soluble in tetrahydrofuran, a solvent for polyphenyls. The dissolved portion was mainly isopropyl diphenyl, the heat-transfer medium with which the shipping casks were filled, and only a trace of high-boiler products was found. The major metallic constituent of the insoluble portion was iron, and X-ray diffraction indicated the presence of Fe_3C and FeC.

The many variables in the reactor history prevented isolation of the factors that caused the heavier fouling of the element with the longer exposure, and evaluation of the deposits was complicated by the contact with isopropyl diphenyl in the shipping casks. Possible contributing factors were the higher exposure, higher high-boiler content of the coolant, higher impurity level of the coolant, and operation of the reactor at higher power levels. The iron present in the film is not believed to have come from the fuel elements but, rather, from iron carbide circulating as an impurity in the coolant.

The damage to fuel element 30 was apparently the result of overheating that occurred when it was improperly installed in the grid plates. Examination in a hot cell showed about the center third of the fuel plates to be extensively damaged, although the areas near the ends were undistorted and free of blockage. Around the center of the element, the fuel plates were buckled away from the side where the cover plate was originally observed to have ruptured. There were scattered massive deposits of a black clinkerlike material between the plates. There appeared, however, to have been space for coolant flow near the edges of even those plates with heavy deposits. Iron was the major metallic constituent of the deposit, and Fe_3C was identified by X-ray diffraction. Less than 1 per cent of the material was soluble in tetrahydrofuran. Since this element had cooled for four months, it was not necessary to supply a heat-transfer medium when it was transferred to the hot cell. In disassembly of the fuel element, most of the plates were found to be stuck together where they had been distorted, and when pried apart, they frequently fractured around the adherent areas. No major disintegration of the element appeared to have occurred until it was forcibly removed from the core.

The following is believed to have been the sequence of events in occurrence of damage to the element. A deposit formed on one side of the fuel element during the period when it was improperly cooled (the thermocoupled fuel plate was on the opposite side of the element and hence did not indicate the increased temperature). The deposit grew, perhaps by absorption and decomposition of coolant in the porous solid, and exerted sufficient force to deform the element. The fuel plates were forced together, became brittle, probably from carburization, and adhered where they touched. It could not be determined at what time the large fission-product release from the stainless-steel element occurred because of the masking effect of the massive release from the failed experimental element. The stainless-steel fuel plates appear to have been remarkably stable, when it is considered that there must have been almost complete loss of coolant flow through large sections of the elements.

Examination of experimental elements HB-1 and HT-1 after removal from the reactor indicated that particulate matter strained out at the inlet end by the aluminum fins restricted flow through the elements and caused them to overheat. The operation of the reactor filtering system up to that time had been unsatisfactory, and the finned fuel elements were the main strainers of particulate matter from the coolant. Partial melting of the aluminum cladding had occurred on several of the plates of HB-1, with consequent release of fission products. The indication of high temperatures by the thermocouples of HT-1, however, resulted in its removal before actual melting occurred.

Radiation Levels and Activity.[12,17] Since the organic coolant does not become activated, any activity in the OMRE coolant is due to impurities, corrosion products, and fission products. Normally no shielding of the plant is necessary, except for protection against radiation coming directly from the core, and access to equipment is possible even during full-power operation. Buildup of activity in the coolant would seriously limit access to the process areas and increase the shielding requirements.

During the early part of the core life, radiation levels near the surface of the 10-in. main circulating pumps were 50 to 60 mr/hr while the reactor was operating at a power level of 6.0 Mw, and the specific activity of the coolant was approximately 0.1 $\mu c/cm^3$. Most of the activity was from activation of impurities in the

form of rust, metal filings, or welding slag which were introduced with the coolant or came from the reactor system. The major activities observed were Mn^{56}, Fe^{59}, P^{32}, and S^{35}. The activity levels later increased by an order of magnitude from the pickup of inorganic particulate matter by the coolant. This material is believed largely to have come from the expansion tank through which circulation was begun in July. Most of the activity was removable from the coolant system by concentration in the high-boiler residue.

Failure of experimental fuel element HB-1 resulted in the release of approximately 1000 curies of activity to the coolant and the gas blanket. The fission products were completely contained in the system, and there was no release to the atmosphere. The radiation level 1 ft from the 10-in.-diameter coolant pipe was 0.04 r/hr prior to failure of the element. The activity level rose to 10 r/hr just after the release, fell to 2.4 r/hr 1 hr later, and was down to 0.05 r/hr 24 hr later. An analysis of the coolant[15] indicated that only fission products which are volatile or have volatile precursors, such as xenon, krypton, rubidium, iodine, barium, lanthanum, and tellurium, were released from the damaged element. A special analysis for zirconium and niobium was negative. Xenon was the only nuclide detected in the blanket gas. The iodine reacted chemically and remained with the coolant. Fission-product activity was removed from the coolant by the purification system, and the blanket gas containing xenon was vented to the atmosphere under controlled conditions. After a few days I^{131}, Ba^{140}, and La^{140} were the only fission products remaining in the coolant in any significant quantity.

Component Performance.[17] The performance of OMRE components has, in general, been very good. The valves and pumps, which mainly are standard oil-refinery items, have given excellent service. There was difficulty initially in getting leaktight flange joints, but this was solved by the substitution of soft iron gaskets for the original spiral asbestos and metal gaskets. Slight leakage from valve stems of the larger valves and from mechanical pump seals has been tolerated because catching the low-vapor pressure coolant in open buckets has worked satisfactorily.[16]

The self-cleaning disk type filter originally installed in the bypass loop proved to be unsuitable because the disk would bind and stop the self-cleaning action when the temperature reached 400°F. The original filter has been replaced with two sintered stainless-steel filters. The importance of effective filtration was demonstrated by the plugging of the coolant passages in the experimental elements.

Although most of the system is made of carbon steel (exceptions are the reactor vessel of mild alloy steel and the stainless-steel-clad fuel elements), there has been no indication of significant corrosion. The particulate matter found in the coolant is believed to have been in the system originally or to have come from the coolant drums. The former source is now avoided by evaporating the coolant into the system.

The control-rod system of the OMRE has operated satisfactorily since early 1957.

Experience with Core 2

Operating experience with core 2 has been reported only in bimonthly progress reports.[19–22] The most recent report covers the period extending through April 1960. Up to this time the second core had accumulated an exposure of 904 Mwd, generally at a high-boiler concentration of 30 per cent. The use of Santowax OMP for coolant makeup was begun in December 1959.

Coolant decomposition rates, reactor stability, and general operating characteristics have been similar to those experienced with the first core loading. Although the out-of-core heat-transfer characteristics were the same in both periods of operation, the fuel-element heat-transfer characteristics of core 2 are stated[22] to have been somewhat inconsistent with previous experience. The nature of the difference is not described, but mention is made of need for additional precision instrumentation (see reference 23 for a discussion of errors in the measurement of fuel-plate temperatures). Evaluation of the data was continuing at the time of the most recent report.

Three fuel elements had been removed from core 2. The first two, removed routinely for hot-cell examination, were coated with a flaky and irregular dark film averaging 0.001 to 0.002 in. in thickness. Selected fuel plates examined for dimensional stability were found to be distorted in a wavelike shape, with a wave

length along the longitudinal center line of 5 to 6 in. and a maximum deviation from straightness of 0.06 to 0.08 in. The distortion was tentatively ascribed to thermal stress. Aside from this waviness, the fuel elements have continued to show good mechanical and irradiation stability.

The third fuel element was removed after 904 Mwd because the outlet temperature in its hottest channel had been steadily increasing. On disassembly, it was found that flaky material, similar in appearance to that found in the first two elements, had collected and filled the 0.134-in. coolant channels in some areas. No conclusion as to the cause of this particular deposit had been reached at the time the report[22] was written. (M. W. Rosenthal)

Action on Reactor Projects by Licensing and Regulating Bodies

In the course of securing final approval for the operation of a power reactor, the applicant must clear several hurdles designed to assure the AEC, and thus the public, that the safety of the public has been adequately considered in a responsible manner. The procedure involves two major steps: (1) securing a construction permit and (2) securing an operating permit, as described in Title 10, *Code of Federal Regulations*.

The processes involved in obtaining a construction permit and in obtaining an operating license require the preparation of a hazards summary report which must be reviewed and approved by both the Division of Licensing and Regulation and the ACRS. As the final step before each license is issued, a public hearing is held. As a consequence of these various administrative reviews, any given reactor may be involved in reviews for a period of years. These reviews frequently result in modification of the reactor design or operation to enhance safety and, in any case, constitute the precedents for "adequate" safety.

Most power reactors proposed for construction and operation in this country are currently involved in some stage of the review process, as indicated in Table VI-1. The reactors discussed below have passed some milestone in the process during the quarter covered by this Review.

Enrico Fermi Atomic Power Plant (Docket F-16)

The Enrico Fermi reactor is a 300-Mw fast breeder reactor which the Power Reactor Development Company proposes to construct at Lagoona Beach, Mich. The hearing on the construction permit was reviewed in the March 1960 issue of *Nuclear Safety*,[24] and in the September 1960 issue of *Nuclear Safety*,[25] the decision of the U. S. Court of Appeals for the District of Columbia setting aside the construction permit for the reactor was reported. The Commission wished to appeal this two-to-one decision and petitioned the U. S. Court of Appeals for a rehearing before the Court *en banc*.[26] After subsequently reviewing statements by the various unions asking that the Court deny the petition for review of its decision and the AEC rebuttal, the Court, on July 25, turned down the request.[27] The AEC then placed its case before the U. S. Supreme Court, where the case now rests.[28] Meanwhile, construction of the facility will apparently continue until the appellate processes are completed.

Florida West Coast Nuclear Power Plant (Docket 50-152)

The Florida West Coast Nuclear Group submitted to the Commission, late in 1959, an application for a construction permit, together with a preliminary hazards summary report.[29] They proposed to build and operate a 153-Mw(t) CO_2-cooled D_2O-moderated reactor in Polk County, Fla. The reactor[29] would have a tubular core section immersed in D_2O. The CO_2-cooled fuel elements of natural uranium would be located within the tubes.

On July 8, 1960, however, the group advised the Commission that no further action on the pending application should be taken.[30] The letter to the Commission stated that the group now wishes to employ beryllium-clad rather than stainless-steel-clad fuel elements in the first core loading and that it is necessary to resolve technical problems concerning this use of beryllium before proceeding with construction of the prototype plant. It is not clear how much the plant will be delayed, but it is apparent that the projected power costs were not sufficiently attractive to warrant proceeding on the basis of the stainless-steel-clad fuel elements. Arrangements are now being made[31] concerning the reoriented research program,

Table VI-1 CALENDAR OF LEGAL STEPS IN LICENSING U. S. POWER REACTORS

Reactor information							Construction permit							
Code No.	Name, operator, and location	Reactor type	Max. power Mw(t)	Max. power Mw(e)	Hazards report No.	Hazards report Date	Findings of ACRS-HEB or AEC staff	Findings of hearing examiner	Permit No.	Permit Date	Permit Term	Schedule of completion date	Additional information	Operating license (and other information)
PP-4	BONUS, Puerto Rico Water Resources Authority, Punta Higüero, Puerto Rico	Boiling H_2O nuclear superheater	50	173	PRWRA-GNEC-2	12-21-59	3-14-60, ACRS, favorable; 6-60, AEC, favorable	6-28-60, favorable	Not required					
50-144	CVTR, Carolinas-Virginia Nuclear Power Associates, Parr, S. C.	Pressurized D_2O moderator and coolant	605	17	CVNA-27 (3 vols.)	7-15-59	12-11-59, AEC, favorable	4-12-60, favorable	CPPR-7	5-4-60	10 yr	1-1-62 to 12-31-62		
50-155	Big Rock Point, Consumers Power Co., Charlevoix County, Mich.	Boiling H_2O	240	75	Not numbered	1-60	4-18-60, ACRS, favorable	4-18-60, favorable	CPPR-9	5-31-60	40 yr	7-1-62 to 7-1-63		
50-10	Dresden, Commonwealth Edison Co., Grundy County, Ill.	Boiling H_2O	630	192 gross, 180 net	Not numbered (4 vols.) Summary	Through 9-57 1-6-59			CPPR-2	5-3-56	40 yr	3-1-60 to 9-30-60	Construction began 3-57, completed 9-59	License No. DPR-2 for half power is dated 9-26-59 and expires 12-10-59; license for temporary full power is dated 6-9-60
PP-1	Elk River, Rural Cooperative Power Associates, Elk River, Minn.	Boiling H_2O	58	22	Not numbered (rev.) Suppl. 1	3-12-59 5-25-59		1-19-60, favorable; 4-60, favorable	Not required					
F-16	Enrico Fermi, Power Reac-	Fast-neutron Pu breeder,	300	104 gross, 94 net	APDA-108 APDA-117	9-1-55 10-15-56			CPPR-4 Reaffirmed 12-10-58	8-4-56	25 yr	12-15-59 to 12-	Construction halted by	

	Power Associates, Elk River, Minn.				Suppl. 1	6-25-59		favorable				
F-16	Enrico Fermi, Power Reactor Development Co., Lagoona Beach, Mich.	Fast-neutron Pu breeder, Na cooled	300	104 gross, 94 net	APDA-108 APDA-117 APDA-115 APDA-124 Exhibits through No. T18	9-1-55 10-15-56 11-1-56 1-59 Through 9-10-59			CPPR-4 8-4-56 25 yr Reaffirmed 12-10-58 Reaffirmed 5-26-59		12-15-59 to 12-15-60	Construction halted by court ruling 6-10-60 (appealed)
	EGCR, Tenn. Valley Authority, Oak Ridge, Tenn.	He cooled, graphite moderated; fuel testing	84	25	ORO-196	5-19-59	5-59, ACRS, favorable		Not required		12-31-62	Construction approved and in progress
50-152	FWCNG, Florida West Coast Nuclear Group, Polk County, Fla.	D_2O moderated, CO_2 cooled	153	50	GNEC-115 (rev.) (3 vols.)	12-7-59	Petition pending					Restudying costs
PP-3	HNPF, Consumers Public Power District, Hallam, Nebr.	Graphite moderated, Na cooled	240	75 gross	NAA-SR-3379 CPPD-25 NAA-SR-4504	No date 9-22-55 No date	7-25-59, ACRS, favorable; 2-8-60, ACRS, favorable; 4-29-60, AEC, favorable	6-6-60, favorable	Not required		7-62	Construction authorized effective 6-28-60
50-133	Humboldt Bay, Pacific Gas & Electric Co., Humboldt Bay, Calif.	Boiling H_2O, vapor suppression (safety)	163	60 gross	Not numbered	4-15-59	Petition pending					Awaiting hearing on construction permit
50-3	Indian Point, Consolidated Edison Co. of New York, Buchanan, N. Y.	Pressurized H_2O; Th converter	585	275 gross 255 net	Exhibits through L-4 Exhibits through K-4 Exhibit K-5 (Rev.1)	Through 1955 Through 1958 1-60			CPPR-1 5-3-56	40 yr	10-1-59 to 10-1-60	

(Table continues on next page.)

Table VI-1 (Continued)

Code No.	Name, operator, and location	Reactor type	Max. power Mw(t)	Max. power Mw(e)	Hazards report No.	Hazards report Date	Construction permit: Findings of ACRS-HEB or AEC staff	Construction permit: Findings of hearing examiner	Construction permit: Permit No.	Construction permit: Permit Date	Construction permit: Permit Term	Schedule of completion date	Additional information	Operating license (and other information)
	N.S. *Savannah* Maritime Administration, U. S. Dept. of Commerce	Pressurized H_2O; special containment	69	22,000 shp	ORNL-2867 BAW-1164 York site	3-21-60 3-60 5-60			Not required				Unique legal status; Presdential recommendation; Congressional authorization; under construction at Camden, N. J.	Critical tests and nuclear-powered trial runs scheduled in 1961
50-130	Pathfinder, Northern States Power Co., Sioux Falls, S. Dak.	Boiling H_2O nuclear superheater	203	66 gross, 62 net	ACNP-5905 ACNP-5917	3-10-59 8-31-59	3-60, ACRS, favorable	4-60, favorable	CPPR-8	5-12-60	40 yr	1-1-62 to 7-1-62		
	Peach Bottom, Philadelphia Electric Co., Conowingo Pond, York County, Pa.	High temperature: unclad fuel; gas cooled; graphite moderated	115	40	Not numbered	12-59	3-14-60, ACRS, favorable							
	Piqua OMR, City of Piqua Municipal Power Commission, Piqua, Ohio	Organic moderated, organic cooled	46	114	NAA-SR-3100SP NAA-SR-3279 NAA-SR-3575 NAA-SR-4048	9-28-58 No date 4-13-59 7-1-59	7-25-59, ACRS, favorable	1-27-60, favorable					Construction started in May 1955	Operating at full power since 12-23-57; new seed element authorized; critical 4-12-60, on line 5-6-60
	PWR, Duquesne Light Co., Shippingport, Pa.	Pressurized H_2O	231	68 gross, 60 net	WAPD-SC-547 WAPD-SC-541 Other supplements	6-57 9-57 11-57								
60-146	Saxton, Saxton Nuclear Ex-	Pressurized H_2O	20	5 net	Not numbered	No date	1-7-60, ACRS,	1-21-60, favor-	CPPR-6	2-11-60		12-1-61 to 12-		License is to expire 2-11-66 or five

					Other supplements	11-57									
50-146	Saxton, Saxton Nuclear Experimental Corp., Berks County, Pa.	Pressurized H_2O	20	5 net	Not numbered	No date	1-7-60, ACRS, favorable	1-21-60, favorable	CPPR-6	2-11-60		12-1-61 to 12-1-62			License is to expire 2-11-66 or five years after criticality, whichever is earlier
50-18	VBWR, General Electric Co., Pleasanton, Alameda County, Calif.	Boiling H_2O test	20	3	GEAP-069 SG-VAL-2 (2nd ed.)	10-17-55 5-8-58			CPPR-3	5-14-56	7 yr	6-1-57 to 2-28-58			License No. DPR-1 is dated 8-31-57 and expires 5-14-63; operating since 9-57; there are more than 40 amendments to this license
50-29	YAEC, Yankee Atomic Electric Co., Rowe, Mass.	Pressurized H_2O	392	145 gross, 136 net	Not numbered	No date		CPPR-5	11-4-57			12-59 to 6-61			ACRS findings on 1-30-60 and 5-9-60 were favorable; findings of the hearing examiner were favorable on 5-17-60
	SSPWR	Small-sized pressurized H_2O	60	165 net	Site report Suppl. 1	2-8-60 3-10-60	Petition pending						Decision on location postponed for additional information		

NOTE: BONUS = Boiling Nuclear Superheater; CVTR = Carolinas-Virginia Tube Reactor; EGCR = Experimental Gas-Cooled Reactor; FWCNG = Florida West Coast Nuclear Group; HNPF = Hallam Nuclear Power Facility; PWR = Pressurized-Water Reactor; VBWR = Vallecitos Boiling-Water Reactor; YAEC = Yankee Atomic Electric Co.; SSPWR = Small-Sized Pressurized-Water Reactor.

which is expected to be along the lines of the third round of the Power Demonstration Reactor Program under which AEC supports the research and development and the utility companies build and operate the plant.

Vallecitos Boiling-Water Reactor

(Docket 50-18)

The Vallecitos Boiling-Water Reactor (VBWR)[32] is a boiling-water reactor, initially licensed and operated at 20 Mw, which has been in operation since 1957. There have been over 40 amendments to the original application, most of which have been reviewed and accepted by the Division of Licensing and Regulation without recourse to the processes involving the ACRS and a public hearing. However, Amendment 41 summarizes all pertinent and required information on the VBWR, and it was therefore deemed desirable that the ACRS review it and Amendments 42 and 43, which were issued shortly thereafter. As noted in the June 1960 issue of *Nuclear Safety*,[33] the ACRS held that the plant could be modified as proposed without undue hazard to the health and safety of the public. Following this review, a public hearing was scheduled at which related issues involving the reporting of incidents and the AEC inspection were discussed, as well as the amendments in question.[34] The hearing examiner subsequently ordered[35] that Amendments 41, 42, and 43 be accepted and also that the license be amended to require that all changes be reported to the Commission for prior approval. (A similar decision had previously been rendered in the case of the Dresden reactor.[36]) The previous license had, with the concurrence of the AEC Division of Licensing and Regulation, permitted the manager–reactor operator to make certain changes which he deemed to be no more hazardous than already allowed in the final hazards summary report. The examiner further specified that the licensee be required to report any unsafe condition observed in reactor operation and all scrams of the reactor, whether emergency or not. Commenting upon the testimony of an AEC Inspection Division witness, who had not been able to make a recent inspection of the facility, the examiner urged the continuation of direct evidence by Inspection Division personnel at licensing proceedings.

Dresden Nuclear Power Station

(Docket 50-10)

The Dresden Nuclear Power Station of the Commonwealth Edison Company has approached the design operating power level of 630 Mw(t) in a number of discrete steps, beginning with the initial authorization for operation at 1 Mw(t) and the subsequent authorization for operation at 315 Mw(t).[37] As noted in the September 1960 issue of *Nuclear Safety*,[38] the ACRS concluded that there was no reason not to continue the approach to full power, and consequently a hearing was held. At the conclusion of this hearing,[39] it was ordered that a license be issued authorizing the "increase of power level operation... up to 630 Mw(t) from 315 Mw(t) but not to operate the reactor in excess of this amount or at a steady-state power level of 630 Mw(t) without further order from the Commission," and that, "a further public hearing be held with regard to a report on rated power tests within 90 days..." The AEC staff had previously proposed[40] that Commonwealth Edison be granted an interim license which would have permitted operation at 630 Mw(t) for three or four months, after which time consideration would be given to the issuance of a 40-year license. It is not clear why the authorization which would permit transient experiments up to 630 Mw(t) and presumably steady-state operation at less than 630 Mw(t) was safer than one which would also permit steady-state operation at 630 Mw(t). Commonwealth Edison expressed objection to both the staff and the hearing examiner's proposals. However, the novel aspects of the reactor control features, the lack of evidence as to the long-term dependability of the in-core ion chambers at the design power level of 630 Mw(t), and the fact that the program to train operating personnel is not now complete compromised their objections. The reactor has since operated satisfactorily[40] up to 630 Mw(t), and Commonwealth Edison has applied for another hearing, as specified in their interim license, to consider the issuance of a 40-year operating license.[41]

Yankee Nuclear Power Station (Docket 50-29)

The Yankee Nuclear Power Station has a pressurized-water reactor that is designed to operate at 485 Mw(t). Following a hearing at which the technical specifications were approved

and the review of Amendments 19 and 20 by the ACRS, as described in the September 1960 issue of *Nuclear Safety*,[42] a hearing extending over several weeks was held to consider the issuance of an operating license. In the course of the hearing, the proposed terms of the license, the filing of late amendments to a reactor project, and who was to start up the reactor were among the more important items relative to safety which were reviewed.[43]

The Company requested a 40-year license for operation at 485 Mw(t), with a review of operations above 392 Mw(t) after 500 hr of operation at that level, and contended that (1) the filing of late amendments which tend to enhance the safety of the facility should not be cause for delay in the licensing procedure and that (2) the proposed startup of the facility by the operator (Yankee) rather than the manufacturer (Westinghouse) constituted good practice. The Yankee position on late amendments was subsequently supported[44] by a memorandum of law submitted to the AEC, which noted that the Atomic Energy Act did not contemplate license amendments and that the AEC was at liberty to devise a licensing procedure which would carry out the purpose of the Atomic Energy Act and its amendments. In order to avoid overloading the ACRS and the hearing procedure, the Yankee memorandum suggested the general adoption of a procedure in which the HEB may approve an amendment if, in their opinion, it does not involve any substantial change in the hazards previously analyzed. With regard to startup of the facility by the operator rather than the manufacturer, as had been the case at Dresden, Yankee contended that they had been able to build up a responsible, trained operating organization which was as qualified as any that would have been drawn from the manufacturer.

The AEC staff expressed satisfaction with the Yankee plant but recommended that, until additional information concerning operating experience is available, the license permit operation up to 392 Mw(t). It was stated that after operation for some time at 392 Mw(t), a further hearing would be held with regard to full-power operation. The AEC staff also stated that the Commission had not yet issued rules relating to when applications for license amendments will be submitted to public hearings and objected to the specification of such procedures in a license, as proposed by Yankee.

By mid-July 1960 the construction of the facility was completed, and the preoperational tests of the reactor were finished "with extremely satisfactory results." The hearing examiner subsequently issued[45] an "order authorizing the initial fuel loading, startup, low-power testing, and related operations of the Yankee reactor up to and including a power level of 5 Mw(t)." Yankee had proposed such an interim low-power license in order to avoid delay in startup. This license was effective immediately, as per a Commission ruling[46] of May 18, which stated that "at the discretion of the Presiding Officer any intermediate decision and order for the issuance of a license... may provide that such decision and order shall become effective immediately upon issue" subject to certain conditions.

Some three weeks after the issuance of the interim low-power license, Yankee Atomic Electric Company received, by order of the hearing examiner, a license authorizing the operation at power levels[47] not to exceed 392 Mw(t). This license was to expire on the effective date of a final decision by the Commission following a public hearing on the operation at 392 Mw(t).

Humboldt Bay Power Plant (Docket 50-133)

The Humboldt Bay Power Plant, which the Pacific Gas and Electric Company proposes to construct on Humboldt Bay, south of Eureka, Calif., is unique in that it is the first and, to date, the only nuclear power plant which would employ pressure-suppression containment. Proceedings leading to the issuance of a construction permit for this facility were postponed, as described in the September 1960 issue of *Nuclear Safety*,[48] principally because of uncertainties regarding the adequacy of the proposed containment. The Company had embarked upon an experimental program designed to demonstrate the efficiency of the proposed pressure-suppression containment. Sufficient additional information was obtained, as reported in Amendments 7 and 8, to enable the ACRS, following its June meeting, to write:[49] "It appears, from the actual test and on the basis of the reported series of measurements, that a satisfactory suppression system can be designed for this reactor." It was also stated that "the Committee believes that baffles in the pool between each vent pipe would make proof test and full-scale conditions comparable." Although

the ACRS concluded that the proposed suppression system could adequately protect those parts of the primary system housed within the dry well (which includes the pressure vessel and approximately 20 per cent of the primary piping), it was concerned that the major portion of the primary piping is outside the dry well and is uncontained against loss of coolant in the event of a pipe rupture and failure of the isolation valve. The ACRS letter expressed the belief "that double isolation valves will provide adequate protection and that as much as possible of the primary piping system outside of the dry well should be shrouded and the shroud vented to the pressure-suppression system."

With these modifications the Committee concluded "that a 200-Mw(t) boiling-water reactor of the design and features proposed can be adequately contained and that it may be constructed with reasonable assurance that it can be operated with the proposed pressure-suppression system at the site selected without creating undue hazard to the health and safety of the public."

There are, nevertheless, several design features that the applicant is still evaluating. Among these features is the Zircaloy-2 fuel-element cladding, where some concern exists regarding self-propagation of small defects but where stainless steel could be substituted. Another is the control-rod system, which adds reactivity if a rod falls downward under the influence of gravity. Consequently, special reliability of the rod-positioning devices and rod-position indication is required.

The Committee's reservations regarding the containment provisions for the primary piping external to the dry well were resolved by the applicant's proposal to install isolation valves in tandem outside the dry well. These valves will be located immediately outside the dry well, and the first valve will connect with a shroud tube that is an extension of the dry well. The ACRS, at its July 1960 meeting, concluded that[50] this proposed arrangement would meet the recommendation contained in its previous letter. Following this review, a hearing on the application for the construction permit was set for August 24.

Hallam Nuclear Power Facility (Docket PP-5)

The Consumers Public Power District proposes to construct and operate a 240-Mw(t) sodium-cooled graphite-moderated reactor at the Hallam Nuclear Power Facility (HNPF) in southeastern Nebraska. The ACRS review and testimony at the hearing on the application for a construction permit were discussed in the September 1960 issue of *Nuclear Safety*.[51] In June, the hearing examiner authorized the issuance of a provisional permit for the construction of the facility. The authorization is described as provisional to the extent that an authorization to operate the facility will not be issued unless a final safeguards report is filed and the Commission, after a further hearing before operation, finds that the final design provides reasonable assurance that the health and safety of the public will not be endangered by operation of the facility.[52]

Boiling Nuclear Superheater Power Station (Docket PP-4)

Authorization for the construction of the Boiling Nuclear Superheater (BONUS) Power Station at Punta Higüero, Puerto Rico, was granted by the hearing examiner following the recent hearing in Puerto Rico. The ACRS and HEB positions on the facility were reviewed in the September 1960 issue of *Nuclear Safety*.[53] At the hearing,[54] four issues were cited:

1. Whether there is information sufficient to provide reasonable assurance that a nuclear reactor of the general type proposed can be constructed and operated at the proposed location without undue risk to the health and safety of the public;
2. Whether there is reasonable assurance that technical information required to complete the safety analysis of this facility will be supplied;
3. Whether General Nuclear Engineering Corporation is technically qualified to design the reactor and whether Maxon Construction Company is technically qualified to construct the reactor; and
4. Whether construction of the reactor will be inimical to the common defense and security or to the health and safety of the public.

The examiner found that:

> Evidence has been adduced to support the affirmative of the first three issues specified for consideration by the Commission. Respecting the fourth issue, there is sufficient evidence for the purposes of this construction permit to conclude in the negative for the fourth issue. When the proceeding for the consideration of evidence to justify operating the proposed reactor is convened, after its construction is completed, the first two [issues] and the last issue, among others, will be re-examined in the light

of the further and additional evidence expected to be available at that time.

Improved-Cycle Boiling-Water Reactor

The Improved-Cycle Boiling-Water Reactor (ICBWR) is a 50-Mw(e) boiling-water reactor which is to be built by General Electric under contract to the AEC. The "improved cycle" refers essentially only to modifications in the steam-generation equipment, and the proposed reactor does not differ significantly from previous boiling-water reactors. The proposed location of this reactor at San Franciscoquito Canyon, Los Angeles County, Calif., was discussed by the ACRS in its letter of June 8 and elaborated upon in its letters of June 27 and July 25. In its first letter[55] the Committee noted that detailed environmental data stressing the atmospheric dispersion characteristics of the site should be obtained but concluded that the site "is suitable for a 50-Mw(e) boiling-water reactor which followed existing technology... [but] has doubts whether this site can be safely expanded into a large power complex." On request of clarification from the AEC, the ACRS stated[56] that the environmental data must be obtained for the specification of necessary engineered safety features of the facility and not that the ACRS doubts the suitability of that site for that reactor. With regard to the proposition of following existing technology, it was noted that the ICBWR would fall within existing technology if the applicant could show that the steam-generation equipment modifications have no significant effect on the nuclear safety of the reactor.

Nuclear Merchant Ship Savannah

The hazards evaluation of the N.S. *Savannah* was reviewed in the June 1960 issue of *Nuclear Safety*.[57] Although the N.S. *Savannah* is Commission-owned and therefore not subject by law to normal review channels, the N.S. *Savannah* has been (and still is) subject to ACRS review, and it is now contemplated that the safety of the nuclear vessel will be further subjected to scrutiny at a public hearing following satisfactory completion of ACRS review. Much additional information has been obtained, and, at the request of the Division of Licensing and Regulation, another review[58] was held by the ACRS in July in order to provide a forum for discussion among the applicants, ACRS, and the AEC staff that would expedite the final review. Despite the additional information, the ACRS was unable to give the Commission the desired assurance of the probable safe operation of the reactor plant because all the necessary information was not available. Containment stress calculations and an improved ventilation system were described, but the design and efficiency of the filter system are yet to be established. The major areas of concern with this reactor are now associated with components and critical testing, the startup procedures, and the adequacy of the startup crew. Despite the tight schedule proposed by the Maritime Administration, the ACRS urged that an unusually cautious schedule be adopted which will guarantee ample time for training of the operating team. (W. B. Cottrell)

Safeguards Reports and Selected Reading

The recently issued safeguards reports and selected literature pertaining to hazards and safety of reactors are listed below for reference. Because of similarity of many reactors, in particular the research reactors, the list is not intended to be all-inclusive.

1. D. F. Hanlen et al., Safety Report for the Critical Reactor Experiment Facility, Report WCAP-1316, Oct. 1, 1959, and Report WCAP-1316(Add. 1), Jan. 15, 1960, Westinghouse Electric Corp., Atomic Power Dept.

2. J. Muraoka, PRTR Hazard Analysis for Various Mechanical Failures, USAEC Report HW-60963(Rev.), Hanford Atomic Products Operation, July 6, 1959.

3. Georgia Institute of Technology, Safeguards Report for the Georgia Tech Research Reactor, Report NP-8350, January 1960.

4. A. F. DiMeglio and J. O. Elliot, Proposal for 1000-kw Operation of the NRL Research Reactor, Report NRL-5358, Naval Research Laboratory, June 19, 1959.

5. F. D. Anderson, SNAP 2 Safety Evaluation Analysis, USAEC Report NAA-SR-4714, Atomics International, Jan. 1, 1960. (Classified)

6. G. P. Dix, Jr., Preliminary Operational Hazards Summary Report for the Task 2 Thermoelectric Generator, USAEC Report MND-P-2184, Martin Co., Nuclear Div., December 1959. (Classified)

7. Hahn-Meitner-Institut für Kernforschung, Supplement to the Safety Report for the Berlin Research Reactor BER, Report HMI-B-6, September 1959 (in German).

8. A. W. Klement, Jr., comp., A List of References on Cesium-137, USAEC Report WASH-1004(Rev.), May 4, 1959.

9. A. W. Klement, Jr., comp., A List of References on Iodine-131, USAEC Report WASH-1003(Rev.), Apr. 15, 1959.

10. A. W. Klement, Jr., comp., A Selected List of References on Radioactive Fallout, USAEC Report WASH-1005(Rev.), Mar. 3, 1959.

11. H. E. Voress, comp., Whole Body Counters: A Literature Search, USAEC Report TID-3543, January 1960.

12. M. Bloomfield and F. G. Bennett, Reactor Kinetics: A Bibliography, USAEC Report NAA-SR-3808, Atomics International, Nov. 1, 1959.

13. T. F. Davis, comp., Radioactive Waste Processing and Disposal: A Bibliography of Selected Classified Report Literature, USAEC Report TID-3069, October 1959. (Classified)

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2. Letter from Harold Price, Director, Division of Licensing and Regulation, to E. T. Morris, General Manager, Westinghouse Test Reactor, Sept. 7, 1960.
3. Westinghouse Electric Corp., Bettis Atomic Power Laboratory, Final Safety Report for the Westinghouse Testing Reactor, Report WCAP-369(Rev.), August 1958.
4. J. A. Cox, Operations Division Quarterly Progress Report, April–June 1958, USAEC Report CF-58-6-122, Oak Ridge National Laboratory, Oct. 13, 1958.
5. *Atomic Energy Clearing House*, 6(28): 32–33 (July 11, 1960).
6. F. T. Binford et al., Comments on WTR Fuel Meltdown, USAEC Report CF-60-5-33, Oak Ridge National Laboratory, May 9, 1960.
7. Private communication from F. T. Binford, ORNL Reactor Operations Division, to R. B. Korsmeyer.
8. *Atomic Energy Clearing House*, 6(33): 7–8 (Aug. 8, 1960).
9. F. R. Bruce et al., Oak Ridge National Laboratory, June 24, 1960. (Unpublished)
10. C. A. Trilling, Operating Experience with the OMRE (May 1957–February 1958), in Proceedings of the SRE-OMRE Forum, USAEC Report TID-7553, pp. 113–150, May 1958.
11. C. A. Trilling, The OMRE — A Test of the Organic Moderator-Coolant Concept, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 9, p. 468, United Nations, New York, 1958.
12. C. A. Trilling, OMRE Operating Experience, *Nucleonics*, 17(11): 113–117, 234 (November 1959).
13. J. H. Walter et al., Evaluation of Irradiated OMRE Fuel Elements First Core Loading, USAEC Report NAA-SR-4641, Atomics International, Mar. 1, 1960.
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p. 518, United Nations, New York, 1958.
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17. Civilian Power Reactor Program. Part III, Status Report on Organic-Cooled Power Reactors as of 1959, USAEC Report TID-8518(Bk.7), 1960.
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21. Atomics International, Bi-Monthly Technical Progress Report, AEC Unclassified Programs, January–February 1960, USAEC Report NAA-SR-5042, Apr. 1, 1960.
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23. S. Sudar, OMRE Fuel Plate Surface Temperature Measurements, USAEC Report NAA-SR-4047, Atomics International, May 1, 1960.
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27. *Atomic Energy Clearing House*, 6(31): 2 (Aug. 1, 1960).

28. *Atomic Energy Clearing House*, 6(33): 19 (Aug. 15, 1960).
29. American Electric Power Service Corp. and General Nuclear Engineering Corp., Application for USAEC Licenses by Florida West Coast Nuclear Group. Part B, Preliminary Hazards Summary Report, AEC Docket No. 50-152, Report NP-8251 (Vols. 1 to 3), Dec. 7, 1959, and Amendment No. 1, Feb. 4, 1960.
30. *Atomic Energy Clearing House*, 6(30): 4 (July 25, 1960).
31. *Atomic Energy Clearing House*, 6(32): 30 (Aug. 8, 1960).
32. L. Kornblith et al., The General Electric Developmental Boiling Water Reactor, Final Hazards Summary Report, Report SG-VAL-2, General Electric Co., Vallecitos Atomic Laboratory, Feb. 15, 1957.
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34. *Atomic Energy Clearing House*, 6(15): 19 (Apr. 11, 1960).
35. *Atomic Energy Clearing House*, 6(25): 17 (June 20, 1960).
36. Hearings on Reactor Projects, *Nuclear Safety*, 1(3): 81 (March 1960).
37. Hearings on Reactor Projects, *Nuclear Safety*, 1(3): 83 (March 1960).
38. Action on Reactor Projects by Licensing and Review Bodies, *Nuclear Safety*, 2(1): 109–110 (September 1960).
39. *Atomic Energy Clearing House*, 6(21): 4 (May 23, 1960).
40. *Atomic Energy Clearing House*, 6(27): 24 (July 4, 1960).
41. *Atomic Energy Clearing House*, 6(32): 6 (Aug. 8, 1960).
42. Action on Reactor Projects by Licensing and Review Bodies, *Nuclear Safety*, 2(1): 113–114 (September 1960).
43. *Atomic Energy Clearing House*, 6(22): 8 (May 30, 1960).
44. *Atomic Energy Clearing House*, 6(26): 22 (June 27, 1960).
45. *Atomic Energy Clearing House*, 6(29): 13 (July 18, 1960).
46. *Atomic Energy Clearing House*, 6(21): 17 (May 23, 1960).
47. *Atomic Energy Clearing House*, 6(32): 7 (Aug. 8, 1960).
48. Action on Reactor Projects by Licensing and Review Bodies, *Nuclear Safety*, 2(1): 111 (September 1960).
49. Letter from L. Silverman to Hon. John A. McCone, June 27, 1960, subject: Humboldt Bay Power Plant.
50. Letter from L. Silverman to Hon. John A. McCone, July 25, 1960, subject: Humboldt Bay Power Plant.
51. Action on Reactor Projects by Licensing and Review Bodies, *Nuclear Safety*, 2(1): 110–111 (September 1960).
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53. Action on Reactor Projects by Licensing and Review Bodies, *Nuclear Safety*, 2(1): 109 (September 1960).
54. *Atomic Energy Clearing House*, 6(27): 28 (July 4, 1960).
55. Letter from L. Silverman to Hon. John A. McCone, June 8, 1960, subject: Improved-Cycle Boiling-Water Reactor.
56. Letter from L. Silverman to Hon. John A. McCone, June 27, 1960, subject: Improved-Cycle Boiling-Water Reactor.
57. Action on Reactor Projects by Licensing and Regulating Bodies, *Nuclear Safety*, 1(4): 93 (June 1960).
58. Letter from L. Silverman to Hon. John A. McCone, July 25, 1960, subject: N.S. *Savannah*.

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